Temporal fluctuations of waves in weakly nonlinear disordered media

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We consider the multiple scattering of a scalar wave in a disordered medium with a weak nonlinearity of Kerr type. The perturbation theory, developed to calculate the temporal autocorrelation function of scattered wave, fails at short correlation times. A self-consistent calculation shows that for nonlinearities exceeding a certain threshold value, the multiple-scattering speckle pattern becomes unstable and exhibits spontaneous fluctuations even in the absence of scatterer motion. The instability is due to a distributed feedback in the system ''coherent wave + nonlinear disordered medium.'' The feedback is provided by the multiple scattering. The development of instability is independent of the sign of nonlinearity.

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I. INTRODUCTION

Scattering of waves in disordered media has proved to be a nontrivial topic possessing intriguing and still not completely understood features [1–5]. Accordingly to the strength of disorder, one observes a variety of propagation regimes ranging from ballistic transport, through single scattering and wave diffusion, to the Anderson localization. In this paper we are interested in the regime of wave diffusion, corresponding to a relatively strong disorder, which is, however, still largely insufficient to bring the system to the localization transition ($kl \gg 1$, where k is a wave number in the medium, and l is a mean-free-path).

It is well known, that multiple scattering of coherent wave in a disordered medium results in a complicated spatial intensity distribution $I(\mathbf{r},t)$ known as a ''speckle pattern.'' The speckle pattern is highly irregular and appears random to the eye. It is now well established that the speckle pattern exhibits large intensity fluctuations [6–8] $\langle \delta I(\mathbf{r},t)^2 \rangle \simeq \langle I(\mathbf{r},t) \rangle^2$, originating from the interference of partial waves arriving at $\mathbf{r}$ with completely randomized phases. Here the angular brackets $\langle \cdots \rangle$ denote ensemble averaging, and $\delta I(\mathbf{r},t) = I(\mathbf{r},t) - \langle I(\mathbf{r},t) \rangle$. Besides, the speckle pattern possesses nontrivial long-range spatial correlation $C_{\delta I}(\mathbf{r},\Delta\mathbf{r}) = \langle \delta I(\mathbf{r} - \Delta\mathbf{r}/2,t)\, \delta I(\mathbf{r}+\Delta\mathbf{r}/2,t) \rangle$ even for $\Delta r > l$. This correlation is due to interaction of diffusing modes [9–12]. If the points $\mathbf{r} \pm \Delta\mathbf{r}/2$ are far enough from the boundaries of the medium, $C_{\delta I} \propto 1/\Delta r$. Recently, it has been shown that in a particular case of a point source of waves embedded inside a disordered medium, there exists an infinite-range contribution to $C_{\delta I}(\mathbf{r},\Delta\mathbf{r})$ originating from scattering events taking place in the immediate neighborhood of the source [13]. This contribution is highly sensitive to the short-distance properties of disorder, as well as to the source size and shape [14].

If the scatterers in the medium are allowed to move, $I(\mathbf{r},t)$ fluctuates with time, and the statistics of these fluctuations is also a subject of active research. A wave propagating in a disordered, multiple-scattering medium undergoes a large number of scattering events, and hence the scattered intensity is highly sensitive to displacements of scatterers [15–17]. Consequently, the decay of the intensity autocorrelation function $C_{\delta I}(\mathbf{r},\tau) = \langle \delta I(\mathbf{r},t)\, \delta I(\mathbf{r},t+\tau) \rangle$ is considerably faster than in the single-scattering case [18–20]. Recently, long-range autocorrelation function of intensity fluctuations has been measured [21], and the existence of the universal conductance fluctuations (analogous to that in disordered conductors) has been demonstrated [22] for optical waves. Theoretical analysis of the temporal correlation function of multiple-scattered waves has been extended to amplifying disordered media [23], as well as to the case of intense incident waves producing flows of scatterers in the disordered medium [24,25]. An additional contribution to $C_{\delta I}(\mathbf{r},\tau)$, originating from scattering in the immediate neighborhood of source and/or detector, decaying much slower than all the previously known contributions, is predicted to exist [14].

High sensitivity of multiple-scattering speckle patterns to scatterer motion gave rise to a new technique for studying the scatterer dynamics in disordered, turbid media, the so-called ''diffusing-wave spectroscopy'' (DWS) [19,26–29]. The latter is now widely applied in concentrated colloidal suspensions [19,26–30], foams [31–35], emulsions [36–38], granular [39–41], and biological [42–44] media. Besides, the DWS has been extended to macroscopically heterogeneous turbid media, providing a tool for imaging of dynamic heterogeneities [45–47] and visualization of scatterer flows [47–49] in the bulk of the medium. A generalization of DWS technique has been also accomplished for anisotropic disordered media [50–52]. Recently, the DWS approach has been extended to nonergodic turbid media [53,54].

The above-mentioned, extensive studies of temporal fluctuations of multiple-scattered waves, as well as the numerous application of DWS, are all restricted to *linear* disordered media. In general, little information is available on the subject of multiple scattering in *nonlinear* disordered media. Meanwhile, the question concerning the way in which the nonlinearity affects the multiple-scattering speckle pattern still remains open and continues to attract research. Consid-

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erable efforts have been made to understand the phenomena of coherent backscattering in disordered media with Kerr-type nonlinearity [55–57]: a narrow dip has been predicted to appear on the top of the backscattering peak. Weak localization effects are shown to exist in the radiation of second harmonic and difference frequency [56,58–60], though their experimental observation failed [61]. Also studied, accounting for disorder, is the optical phase conjugation [62–65]. More recently, correlations in transmission and reflection coefficients of second harmonic waves have been investigated both theoretically and experimentally [66], and the angular correlation functions of fundamental wave in a disordered medium with Kerr-type nonlinearity have been calculated [67]. Despite the fact that theoretical description of wave scattering in nonlinear media is complicated by the simultaneous presence of both disorder and nonlinearity, the standard diagram technique for impurity scattering has been extended to the case of disordered medium with nonlinearity of Kerr type [68,69].

Very recently, it has been shown that the speckle pattern resulting from the multiple scattering of coherent wave in a *nonlinear* disordered medium with Kerr-type nonlinearity, should be extremely sensitive to changes of scattering potential [70], i.e., much more sensitive than the *linear* speckle pattern. This high sensitivity has been explained by the multiplicity of solutions of nonlinear wave equation [70]. The multiplicity of solutions has been then shown to lead to the temporal *instability* of the multiple-scattering speckle pattern in nonlinear medium, resulting in *spontaneous* fluctuations of scattered wave with time [71]. An important prediction of Ref. [71] is that the nonlinearity should exceed some threshold value for the instability to develop. The threshold value is principally determined by the absorption length L_a, or by the sample size L, if $L<L_a$. The striking feature of the established result is that the threshold value of nonlinearity tends to zero in an unbounded medium without absorption. Purely elastic, unbounded nonlinear multiple-scattering systems are therefore always unstable. The physical origin of the instability is easy to understand [71]. Nonlinearity modifies the phases of partial waves propagating in the medium. The phase modifications are proportional to the intensity $I(\mathbf{r},t)$ and affect the mutual interference of partial waves. As it is this interference that is responsible for $I(\mathbf{r},t)$, a sort of feedback establishes in the medium. A small modification of $I(\mathbf{r},t)$ causes modifications of phases of partial waves which, in their turn, produce changes of $I(\mathbf{r},t)$, and so on. It is well known that nonlinear wave systems with sufficiently strong, positive feedback, may become unstable [72,73]. As an example, we cite a family of nonlinear optical systems with two-dimensional feedback [73–76], where spontaneous formation of complicated spatial structures is observed. Despite the absence of disorder, such systems exhibit transition to seemingly chaotic dynamics with increasing nonlinearity [75,76]. An analogy can be drawn between the nonlinear optical systems with two-dimensional feedback and nonlinear disordered media by considering the scattering as a (three-dimensional) feedback mechanism. In the case of disordered media, however, the feedback is of random nature and it is therefore hopeless to expect regular spatial structures to form. Meanwhile, the instability can manifest itself in spontaneous fluctuations of speckle pattern. In order to clarify the issue of instability of speckle patterns in nonlinear disordered media, we consider the following questions:

(a) Can the multiple scattering provide a positive feedback mechanism for waves propagating in a nonlinear disordered medium?

(b) If ''yes,'' how strong should the nonlinearity be for the instability to develop?

A general announcement of our principal answers to the above questions has been given in our recent letter [71]. In the present paper, we discuss and justify the assumptions and approximations underlying our conclusions, provide the missing details of calculations, and give a comprehensive discussion of results. Also developed and discussed is the perturbation approach to the calculation of the temporal autocorrelation function of multiple-scattered wave in a nonlinear disordered medium. It is important that the validity condition of the perturbation theory coincides with the condition for the instability threshold as obtained by using the self-consistent approach. In addition, we give a detailed consideration to an experimentally important case of moving scatterers, when the decrease of the time autocorrelation function is due to a combined effect of spontaneous and scatterer-motion-induced fluctuations of the speckle pattern.

The remainder of the paper is arranged as follows. In Sec. II, we introduce the nonlinear wave equation, and discuss how the path-integral approach can be applied for its analysis. We also formulate the basic models and approximations used throughout the paper. Section III is devoted to linear disordered media. In this section, we provide the expressions for the spatiotemporal intensity correlation functions. Although correlations of multiple-scattered waves in linear media are well studied nowadays, we present their first, to our knowledge, treatment with a simultaneous account for absorption, boundary conditions at the sample surface, and scatterer motion. The results of Sec. III serve as a base for further calculations. In Sec. IV, we present a calculation of dephasing of waves in a nonlinear disordered medium. Our calculation takes into account the fluctuations of the local refractive index due to nonlinear effects, as well as the long-range spatial correlation of these fluctuations. Three ''nonlinear'' contributions to the dephasing are identified in addition to the usual, ''linear'' term originating directly from the motion of scatterers. Further, in Sec. V we develop a perturbation theory for calculation of the temporal autocorrelation function of a multiple-scattered wave, and show its failure at short correlation times, for sufficiently weak absorption. A condition of validity of the perturbation theory is established by comparing the linear and nonlinear contributions to the dephasing found in Sec. IV. Section VI presents an alternative, self-consistent approach to the calculation of the temporal autocorrelation of scattered wave. Development of self-consistent theory requires some additional assumptions, which are also discussed in this section. In Sec. VII, the main results of our self-consistent approach are presented and discussed. The multiple-scattering speckle pattern is shown to exhibit spontaneous fluctuations even in the absence of scatterer motion, which we interpret as a signature of its insta-

bility. A comparison of self-consistent and perturbative results is given, and the condition of the speckle pattern instability is shown to coincide with the condition of validity of the perturbation theory. Finally, concluding remarks are presented in Sec. VIII. In order to maintain the text of the paper readable, we have chosen to collect the technical details of calculations in three appendices. Appendix A is devoted to the derivation of the field-field spatiotemporal correlation function. In Appendix B we compute the spatiotemporal long-range intensity correlation function. Appendix C provides the details of calculations of path distributions $\rho_s(\mathbf{r})$ and $\rho_s(\mathbf{r},\mathbf{r}')$ defined in Sec. IV.

II. WAVE EQUATION AND PATH INTEGRALS

We consider a scalar monochromatic wave of frequency ω propagating in a random medium with Kerr-type nonlinearity. The wave amplitude $\psi(\mathbf{r},t)$ obeys a nonlinear wave equation [77,78]:

$$\{\nabla^2+k_0^2[\varepsilon_0'+i\varepsilon_0''+\delta\varepsilon(\mathbf{r},t)+\varepsilon_2|\psi(\mathbf{r},t)|^2]\}\psi(\mathbf{r},t)=0. \tag{1}$$

Here k_0 is the free-space wave number, $\varepsilon_0=\varepsilon_0'+i\varepsilon_0''$ is the average (complex) dielectric function, $\delta\varepsilon(\mathbf{r},t)$ is the fluctuating part of the dielectric function, and ε_2 is the nonlinear susceptibility [79] (the two latter quantities are assumed to be real). Equation (1) is valid only if $\delta\varepsilon(\mathbf{r},t)+\varepsilon_2|\psi(\mathbf{r},t)|^2$ does not change significantly on the time scale of ω^{-1}. The expression in the square brackets of Eq. (1) can be considered as some ''effective'' dielectric function of the medium. General analysis of Eq. (1) for arbitrary relation between various terms comprising this function constitutes a formidable task, and is not a purpose of this paper. We assume the following hierarchy:

$$\langle\varepsilon_2^2|\psi(\mathbf{r},t)|^4\rangle\ll\langle\delta\varepsilon^2(\mathbf{r},t)\rangle,\quad \langle\varepsilon_2^2|\psi(\mathbf{r},t)|^4\rangle\ll\varepsilon_0',$$
$$|\varepsilon_0''|\ll|\varepsilon_0'|. \tag{2}$$

In other words, we assume that the role of nonlinearity is less significant than that of disorder, and that absorption is weak allowing multiple scattering of waves in the medium. It is then convenient to define the effective refractive index $n_0=(\varepsilon_0')^{1/2}$, the absorption length $l_a=n_0/(k_0\varepsilon_0'')$, and the nonlinear coefficient $n_2=\varepsilon_2/(2n_0)$, which determines the nonlinear correction to the (linear) refractive index of the medium: $n(\mathbf{r},t)=n_0+n_2I(\mathbf{r},t)$, where $I(\mathbf{r},t)=|\psi(\mathbf{r},t)|^2$ is the wave intensity.

In this paper, we study the fluctuations of the solution of Eq. (1) with time t. In a linear medium ($\varepsilon_2=0$), these fluctuations can only be due to random fluctuations of $\delta\varepsilon(\mathbf{r},t)$ with time. The fluctuations of $\psi(\mathbf{r},t)$ are commonly characterized by the autocorrelation function $C_\psi(\mathbf{r},\tau)=\langle\psi(\mathbf{r},t)\psi^*(\mathbf{r},t+\tau)\rangle$. We assume that this autocorrelation function is independent of t, which implies that for fixed $\mathbf{r}$, $\psi(\mathbf{r},t)$ represents a stationary random process (this is obviously true if the sample geometry and the source distribution do not change with time, and if $\delta\varepsilon(\mathbf{r},t)$ is a stationary random process). We take $\delta\varepsilon(\mathbf{r},t)$ to be a Gaussian random field with zero mean and the correlation function $C_{\delta\varepsilon}(\Delta\mathbf{r},\tau)=\langle\delta\varepsilon(\mathbf{r}-\Delta\mathbf{r}/2,t)\delta\varepsilon(\mathbf{r}+\Delta\mathbf{r}/2,t+\tau)\rangle$. For a medium composed of pointlike scatterers undergoing Brownian motion with a diffusion coefficient D_B [18,20],

$$C_{\delta\varepsilon}(\Delta\mathbf{r},\tau)=\frac{4\pi/(k^4l)}{(4\pi D_B\tau)^{3/2}}\exp\left(-\frac{\Delta r^2}{4D_B\tau}\right), \tag{3}$$

where $k=k_0n_0$, and the mean-free-path $l\ll l_a$ is introduced (a weak scattering limit $kl\gg1$ is assumed). A natural time scale for scattering of waves in the medium described by Eq. (3) is set by the characteristic time needed for a scatterer to move a distance of the order of the wavelength: $\tau_0=(4k^2D_B)^{-1}$. From here on, we will be interested in short correlation times $\tau\ll\tau_0$.

In the linear case ($\varepsilon_2=0$), several approaches have been elaborated to analyze Eq. (1). We mention the diagrammatic techniques [80,81], theory of radiative transfer [7], and the method of path integrals [82,83]. The three approaches are known to give equivalent results for C_ψ at $\tau\ll\tau_0$. In the present paper, we adopt the method of path integrals that was originally proposed in the framework of quantum electrodynamics [84], but later has been successfully used in various areas of physics [85], and, in particular, for the analysis of wave scattering problems [82,83]. The method is based on the fact that the solution $\psi(\mathbf{r},t)$ of the wave equation (1) can be written in a form of a functional integral, with integration performed over all possible trajectories (paths) going from the source of waves to $\mathbf{r}$ [82]. Since in the weak scattering limit ($kl\gg1$) different trajectories can be considered independently, it appears that the correlation function C_ψ reduces to the following integral [26–29]:

$$C_\psi(\mathbf{r},\tau)=I_0\int_0^\infty P(\mathbf{r},s)\exp\left[-\frac{1}{2}\langle\Delta\varphi^2(\tau)\rangle_s\right]ds, \tag{4}$$

where I_0 is the average intensity in a nonabsorbing medium, $P(\mathbf{r},s)$ is a weight coefficient of paths of length s, and $\langle\Delta\varphi^2(\tau)\rangle_s$ denotes the squared phase difference $\Delta\varphi(t,\tau)=\varphi(t+\tau)-\varphi(t)$, averaged over various realizations of disorder, and over all possible paths of the same length s. From here on, we denote such an averaging by $\langle\cdots\rangle_s$. Note that $\langle\Delta\varphi(\tau)\rangle_s=0$ for the model of Brownian pointlike scatterers. Meanwhile, [27–29]

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(0)}=\frac{\tau}{\tau_0}\frac{s}{l}, \tag{5}$$

where the superscript (0) denotes the linear case. It is worth noting that $\langle\Delta\varphi^2(\tau)\rangle_s^{(0)}$ does not depend neither on the sample geometry, or on the source and detector positions. Its value is only determined by the scatterer dynamics (through the single-scattering correlation time τ_0), and the path length s. In contrast, $P(\mathbf{r},s)$ can only be calculated if the sample geometry, source distribution, and detector position $\mathbf{r}$ are specified. In what follows, we restrict our analysis to a semi-infinite medium occupying the half-space $z>0$, and illuminated by a plane monochromatic wave incident at $z=0$. For $s\gg l$, $P(\mathbf{r},s)$ becomes [27–29,86]

$$P(\mathbf{r},s)=\left(\frac{3z^2}{4\pi l s^3}\right)^{1/2}\exp\left(-\frac{3z^2}{4sl}-\frac{s}{l_a}\right). \tag{6}$$

Once the field correlation $C_\psi(\mathbf{r},\tau)$ is known, the autocorrelation of intensity fluctuations $C_{\delta I}(\mathbf{r},\tau)=\langle \delta I(\mathbf{r},t)\,\delta I(\mathbf{r},t+\tau)\rangle$ can be found applying the factorization approximation: $C_{\delta I}(\mathbf{r},\tau)=|C_\psi(\mathbf{r},\tau)|^2$ [87].

Combining together Eqs. (4)–(6), one obtains the normalized autocorrelation function of multiple-scattered wave in a semi-infinite disordered medium:

$$g_1^{(L)}(\mathbf{r},\tau)=\frac{C_\psi(\mathbf{r},\tau)}{C_\psi(\mathbf{r},0)}=\exp\left\{-\left[\alpha(\tau)-\frac{l}{L_a}\right]\frac{z}{l}\right\}, \tag{7}$$

where the superscript (L) denotes the linear case, $\alpha^2(\tau)=3\tau/(2\tau_0)+l^2/L_a^2$, and $L_a=(ll_a/3)^{1/2}\gg l$. For the diffusely reflected wave, we assume $z\simeq l$ and get

$$g_1^{(L)}(l,\tau)\equiv g_1^{(L)}(\tau)=\exp\left\{-\alpha(\tau)+\frac{l}{L_a}\right\}. \tag{8}$$

From here on, we will use $g_1(\mathbf{r},\tau)$ to denote the normalized autocorrelation function at a point $\mathbf{r}$ *inside* the medium, while $g_1(\tau)$ — for the normalized autocorrelation function of *diffusely reflected* wave. The superscripts (L) and (NL) will be used to distinguish between linear and nonlinear cases.

Now we turn to the nonlinear medium. Strictly speaking, the method of path integrals cannot be applied for the analysis of Eq. (1), once $\varepsilon_2\neq 0$. The failure of the path-integral technique follows from the fact that this approach relies on the superposition principle, which is not valid for waves in nonlinear media. However, if the nonlinearity is weak [which is ensured by the first two inequalities of Eq. (2)], Eq. (4) is still approximately valid provided that its main ingredients $P(\mathbf{r},s)$ and $\langle\Delta\varphi^2(\tau)\rangle_s$ are computed with account for nonlinear effects. To simplify such a calculation, we assume that the nonlinearity is sufficiently weak to validate the following two assumptions:

(i) Propagation of waves in a weakly nonlinear disordered medium is diffusive with a mean-free-path l unaffected by the nonlinearity. This implies that nonlinear refraction is negligible at distances of order l, and consequently, that $\Delta n^2 kl\ll 1$, where $\Delta n=n_2 I_0$, and I_0 is the average intensity in the absence of absorption. This assumption is an alternative formulation of the fact that the role of nonlinearity is much less significant than that of disorder [see also the first two inequalities of Eq. (2)].

(ii) Intensity of the third harmonic remains always much smaller than the intensity of the fundamental wave. This implies either that $\psi(\mathbf{r},t)$ is considered as a complex quantity [in this case, Eq. (1) is a nonlinear Schrödinger equation, $|\psi(\mathbf{r},t)|^2$ is time-independent, and the third harmonic is not generated at all], or that the medium has a sufficient degree of dispersion for the phase matching condition [77,78] to be violated: $|\Delta k|l\gg 1$ with $\Delta k=k_3-3k$ and k_3 being the wave number at frequency 3ω.

Assumption (i) allows us to consider the path distribution $P(\mathbf{r},s)$ being unaffected by nonlinearity. The only object to be recalculated, accounting for nonlinear effects, is then $\langle\Delta\varphi^2(\tau)\rangle_s$. Before going into an explicit calculation of $\langle\Delta\varphi^2(\tau)\rangle_s$, we devote the next section to a brief derivation of some important results for linear medium.

III. CORRELATIONS IN A LINEAR MEDIUM

As indicated above, we consider a monochromatic plane wave incident at the surface $z=0$ of a semi-infinite medium occupying the half-space $z>0$. The average intensity at $z>l$ can be then found in the diffusion approximation: [7,88] $\langle I(\mathbf{r},t)\rangle=I_0\exp(-z/L_a)$. The spatiotemporal correlation function of the field is given by a solution of the Bethe-Salpeter equation (see Appendix A for details of the calculation):

$$\begin{aligned}C_\psi(\mathbf{r},\Delta\mathbf{r},\tau)&=\langle\psi(\mathbf{r}-\Delta\mathbf{r}/2,t)\,\psi^*(\mathbf{r}+\Delta\mathbf{r}/2,t+\tau)\rangle\\&=I_0\frac{\sin(k\Delta r)}{k\Delta r}\exp\left\{-\frac{\Delta r}{2l}-\alpha(\tau)\frac{z}{l}\right\}.\end{aligned} \tag{9}$$

Let us now consider the correlation functions of intensity fluctuations. In addition to $\delta I(\mathbf{r},t)$, which is the deviation of intensity from its average value, it is convenient to define $\Delta I(\mathbf{r},t,\tau)=I(\mathbf{r},t+\tau)-I(\mathbf{r},t)$, which is the change of the local intensity during the time interval τ. While $\langle\delta I(\mathbf{r},t)\rangle=\langle\Delta I(\mathbf{r},t,\tau)\rangle=0$, the correlation functions $C_{\delta I}(\mathbf{r},\Delta\mathbf{r},\tau)$ and $C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)=\langle\Delta I(\mathbf{r}-\Delta\mathbf{r}/2,t,\tau)\Delta I(\mathbf{r}+\Delta\mathbf{r}/2,t,\tau)\rangle$ for $\Delta r<l$ can be found in the factorization approximation:

$$C_{\delta I}(\mathbf{r},\Delta\mathbf{r},\tau)=|C_\psi(\mathbf{r},\Delta\mathbf{r},\tau)|^2, \tag{10}$$

$$C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)=2[C_{\delta I}(\mathbf{r},\Delta\mathbf{r},0)-C_{\delta I}(\mathbf{r},\Delta\mathbf{r},\tau)]. \tag{11}$$

Both correlation functions (10) and (11) decrease exponentially with $\Delta r/l$, and thus become negligible for $\Delta r>l$. Intensity correlation persists, however, even for two points separated by a distance $\Delta r>l$. This correlation is due to the diffusive nature of wave transport in the medium and can be found either using the Langevin approach [9,11] or applying diagrammatic methods [10,12]. We give the details of calculations in Appendix B, the final results are

$$\begin{aligned}C_{\delta I}(\mathbf{r},\Delta\mathbf{r},\tau)&\\=\frac{3}{(kl)^2}I_0^2\int_0^\infty dK\,K\,Q&\left(K,\sqrt{K^2+l^2/L_a^2},\frac{z}{l},\frac{\Delta z}{l},\alpha(\tau)\right)\\&\times J_0(K\Delta R/l),\end{aligned} \tag{12}$$

$$\begin{aligned}C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)=\frac{6}{(kl)^2}I_0^2\int_0^\infty dK\,K\,\Delta Q&\\\times\left(K,\sqrt{K^2+l^2/L_a^2},\frac{z}{l},\frac{\Delta z}{l},\alpha(\tau),\alpha(0)\right)&\\\times J_0(K\Delta R/l).&\end{aligned} \tag{13}$$

Here we use the cylindrical coordinates: $\mathbf{r}=\{\mathbf{R},z\}$, J_0 is the Bessel function of zeroth order, the function Q is defined in Appendix B, and

$$\Delta Q(\ldots,\alpha(\tau),\alpha(0))=Q(\ldots,\alpha(0))-Q(\ldots,\alpha(\tau)). \tag{14}$$

Due to a rather complicated structure of the function Q (see Appendix B), further calculations can be done only approximately. In the case of $z\pm\Delta z/2,\Delta r\ll l/\alpha(\tau)$, we get

$$C_{\delta I}(\mathbf{r},\Delta\mathbf{r},\tau)\simeq\frac{3}{4}\frac{I_0^2}{(kl)^2}\frac{l}{z_g}\left[\frac{1}{\chi}-\frac{1}{\sqrt{1+\chi^2}}\right]\exp\left[-2\alpha(\tau)\frac{z}{l}\right], \tag{15}$$

where z_g is the geometrical average of z coordinates of the two points for which the correlation is computed: $z_g=\sqrt{z^2-\Delta z^2/4}$ and $\chi=\Delta r/(2z_g)$. For $\alpha(\tau)=0$, Eq. (15) is exact. If $\chi<1$, the correlation behaves essentially as $1/\Delta r$, while for $\chi>1$ it becomes proportional to $z_g^2/\Delta r^3$. For the correlation function of Eq. (13) we find

$$C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)\simeq\begin{cases}4[\alpha(\tau)-\alpha(0)](z/l)C_{\delta I}(\mathbf{r},\Delta\mathbf{r},0), & 2\alpha(\tau)(z/l)\ll 1,\\ 2C_{\delta I}(\mathbf{r},\Delta\mathbf{r},0), & 2\alpha(\tau)(z/l)\gg 1.\end{cases} \tag{16}$$

IV. DEPHASING OF WAVES IN A NONLINEAR MEDIUM

Consider a single wave path of length s going from the source of waves to some point $\mathbf{r}$. The phase acquired by a wave traveling along such a path can be written as

$$\varphi(t)=\int_0^s k_0 n[\mathbf{r}(s_1),t]ds_1, \tag{17}$$

where the integration is along the path, and $n(\mathbf{r},t)=n_0+n_2I(\mathbf{r},t)$. The squared difference $\Delta\varphi(t,\tau)=\varphi(t+\tau)-\varphi(t)$, averaged over various realizations of disorder, and over all possible paths of length s, is found directly from Eq. (17):

$$\langle\Delta\varphi^2(\tau)\rangle_s=\sum_{j=0}^{3}\langle\Delta\varphi^2(\tau)\rangle_s^{(j)}, \tag{18}$$

where the four contributions corresponding to $j=0,\ldots,3$ originate from different physical processes. Below, we give explicit expressions of these terms and discuss their origin.

The first term in Eq. (18), $\langle\Delta\varphi^2(\tau)\rangle_s^{(0)}$ is the linear term given by Eq. (5). The next three terms, namely, the terms corresponding to $j=1$, 2, and 3, are absent in the linear case and only appear because of nonlinear nature of wave interaction with the medium. Explicit expressions for these terms are

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(1)}=\left\langle\frac{2n_2}{n_0 l}\frac{\tau}{\tau_0}\int_0^s\langle I(\mathbf{r})\rangle ds_1\right\rangle_s=\frac{2n_2}{n_0}\frac{\tau}{\tau_0}\langle\langle I(\mathbf{r})\rangle\rangle_s\frac{s}{l}, \tag{19}$$

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(2)}=\left\langle\frac{\pi}{n_0}k_0n_2^2\int_0^s C_{\Delta I}(\mathbf{r},0,\tau)\,ds_1\right\rangle_s=\frac{\pi}{n_0}k_0 l n_2^2\langle C_{\Delta I}(\mathbf{r},0,\tau)\rangle_s\frac{s}{l}, \tag{20}$$

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(3)}=\left\langle k_0^2n_2^2\int_0^s\int_0^s C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)ds_1ds_2\right\rangle_s=(k_0l)^2n_2^2\langle C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)\rangle_s\left(\frac{s}{l}\right)^2. \tag{21}$$

Here the integrations are assumed along wave paths of length $s\gg l$ [in Eq. (21), both integrals are along the same path]. Equation (20) originates from the short-range correlation of intensity fluctuations [see Eqs. (9)–(11)]:

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(2)}=k_0^2n_2^2\langle C_{\Delta I}(\mathbf{r},0,\tau)\rangle_s\int_0^s ds\int_{-l}^{l}d(\Delta s)\times\left[\frac{\sin(k\Delta s)}{k\Delta s}\right]^2\exp\left(-\frac{\Delta s}{l}\right), \tag{22}$$

where the wave path is assumed to be ballistic at distances shorter than l. Equation (22) reduces to (20) for $kl\gg 1$. Next, the term given by Eq. (21) is due to the long-range correlations of intensity fluctuations [see Eq. (13)]. The averages entering into the right-hand sides of Eqs. (19)–(22) are

$$\langle\langle I(\mathbf{r})\rangle\rangle_s=\int d^3\mathbf{r}\rho_s(\mathbf{r})\langle I(\mathbf{r})\rangle, \tag{23}$$

$$\langle C_{\Delta I}(\mathbf{r},0,\tau)\rangle_s=\int d^3\mathbf{r}\rho_s(\mathbf{r})C_{\Delta I}(\mathbf{r},0,\tau), \tag{24}$$

$$\langle C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)\rangle_s=\int d^3\mathbf{r}_1\int d^3\mathbf{r}_2\rho_s(\mathbf{r}_1,\mathbf{r}_2)C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau), \tag{25}$$

where the integrations are over the volume of the disordered medium, and $\mathbf{r}_{1,2}=\mathbf{r}\pm\Delta\mathbf{r}/2$. In Eqs. (23)–(25), $\rho_s(\mathbf{r})$ is the probability density for a path of length s, to pass through the vicinity of $\mathbf{r}$, and $\rho_s(\mathbf{r}_1,\mathbf{r}_2)$ is the probability density for the path to pass consequently through the vicinities of $\mathbf{r}_1$ and $\mathbf{r}_2$. These two ''path distributions'' should be calculated with

account for a particular geometry of disordered sample and source of waves. Once the geometry is fixed, the calculation is straightforward. For the case of a plane wave incident upon a semi-infinite disordered medium, the calculations of $\rho_s(\mathbf{r})$ and $\rho_s(\mathbf{r}_1,\mathbf{r}_2)$ are presented in Appendix C.

Let us discuss briefly the physical origins of nonlinear contributions to the dephasing given by Eqs. (19)–(21). $\langle\Delta\varphi^2(\tau)\rangle_s^{(1)}$ describes the change of the effective wave number in the nonlinear medium: $k=k_0n_0\rightarrow k(\mathbf{r})=k_0[n_0+n_2\langle I(\mathbf{r},t)\rangle]$. This contribution can be either positive or negative, depending on the sign of n_2, but its absolute value is always much smaller than $\langle\Delta\varphi^2(\tau)\rangle_s^{(0)}$, as long as $|\Delta n|\ll n_0$. $\langle\Delta\varphi^2(\tau)\rangle_s^{(1)}$ can therefore only cause a small correction to the linear correlation function. The next contribution, $\langle\Delta\varphi^2(\tau)\rangle_s^{(2)}$ originates from fluctuations of the local intensity, while $\langle\Delta\varphi^2(\tau)\rangle_s^{(3)}$ is due to the long-range correlation of these fluctuations. An important difference between the linear term (5), the first nonlinear term (19), and the two last nonlinear terms (20) and (21), is that the latter terms do not depend explicitly on τ_0. The terms given by Eqs. (20) and (21) are determined by the *intensity fluctuations*, and not by the scatterer displacements. This might seem to be a meaningless statement, as the intensity fluctuations are, in their turn, caused by the scatterer motion. The important point is that the scatterer motion is not the only possible reason for the fluctuations of intensity with time. Weak, spontaneous fluctuations of $I(\mathbf{r},t)$ (due to thermal fluctuations of various parameters, vibrations in the experimental setup, fluctuations of the incident wave, etc.) are inevitable in real physical systems. Equations (20) and (21) provide a mechanism for this spontaneous and generally weak fluctuations to affect the dephasing $\langle\Delta\varphi^2(\tau)\rangle_s$ and, consequently, the temporal correlation function of scattered wave.

V. PERTURBATION THEORY

As stated in the title, the present paper is devoted to *weakly* nonlinear disordered media. We limit ourselves to a weak nonlinearity, as otherwise the problem becomes too involved. Above, we have already mentioned that we assume $\Delta n^2kl\ll 1$, and that this condition allows us to consider the transport of average intensity to remain unaffected by the nonlinearity [assumption (i) of Sec. II]. This allows us to use ''linear'' results, $\langle I(\mathbf{r})\rangle=I_0\exp(-z/L_a)$ and Eqs. (C5) and (C8) of Appendix C, for $\langle I(\mathbf{r})\rangle$, $\rho_s(\mathbf{r})$, and $\rho_s(\mathbf{r}_1,\mathbf{r}_2)$ in Eqs. (23)–(25). It seems then natural to assume that $C_{\Delta I}(\mathbf{r},0,\tau)$ and $C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)$ are also close to their linear values. We can therefore replace these correlation functions in Eqs. (24) and (25) by the expressions found in Sec. III. Then, making use of Eqs. (10)–(13), and performing necessary integrations, we obtain from Eqs. (19)–(21):

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(1)}=2\Delta n\frac{\tau}{\tau_0}\left\{1-H\left(\alpha(0)\sqrt{\frac{s}{12l}}\right)\right\}\frac{s}{l}, \quad (26)$$

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(2)}=2\pi k_0l\Delta n^2\left\{H\left(\alpha(\tau)\sqrt{\frac{s}{3l}}\right)-H\left(\alpha(0)\sqrt{\frac{s}{3l}}\right)\right\}\frac{s}{l}, \quad (27)$$

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(3)}=6\Delta n^2S(\alpha(\tau)\sqrt{s/l},\alpha(0)\sqrt{s/l})\left(\frac{s}{l}\right)^{3/2}. \quad (28)$$

Here $H(x)=\sqrt{\pi}x\exp(x^2)[1-\mathrm{Erf}(x)]$ and $n_0=1$ is assumed for simplicity. The function $S(u,v)$ in Eq. (28) is

$$S(u,v)=9\int_0^\infty dRR\int_0^\infty dKK\int_0^\infty dz\int_0^{2z}d(\Delta z)f(z,\Delta z,R)\times\Delta Q(K,\sqrt{K^2+v^2},z,\Delta z,u,v)J_0(KR), \quad (29)$$

$$f(z,\Delta z,R)=\frac{2z+\sqrt{R^2+\Delta z^2}}{\sqrt{R^2+\Delta z^2}}\exp\left[-\frac{3}{4}(2z+\sqrt{R^2+\Delta z^2})^2\right]-\frac{2z+\sqrt{R^2+4z^2}}{\sqrt{R^2+4z^2}}\exp\left[-\frac{3}{4}(2z+\sqrt{R^2+4z^2})^2\right]. \quad (30)$$

Unfortunately, integrations in Eq. (29) cannot be performed in the general case. We find, however, the following approximate results:

$$S(u,v)\simeq\begin{cases}(u-v), & u-v\leqslant 1,\ v\leqslant 1,\\ 1, & u-v>1,\ v\leqslant 1,\\ (u-v)v^{-3}, & u-v\leqslant 1,\ v>1,\\ v^{-3}, & u-v>1,\ v>1.\end{cases} \quad (31)$$

Here numerical factors of order unity are omitted before each of the four asymptotic expressions.

While approximate, the above results enable one to compute the temporal autocorrelation function $g_1^{(NL)}(\tau)$ of diffusely reflected wave numerically using Eqs. (4), (6), and (18):

$$g_1^{(NL)}(\tau)=W[\langle\Delta\varphi^2(\tau)\rangle_s]/W[0], \quad (32)$$

$$W[\langle\Delta\varphi^2(\tau)\rangle_s]=\int_0^\infty P(l,s)\exp\left[-\frac{1}{2}\langle\Delta\varphi^2(\tau)\rangle_s\right]ds. \quad (33)$$

We present the results of the calculation in Fig. 1 for fixed Δn, k_0l, and the values of the inverse absorption length l/L_a indicated near each curve. The ''linear'' correlation functions, corresponding to the same three values of the absorption length, and to $\Delta n=0$, are shown by dashed lines. For weak absorption ($l/L_a=0$, 10^{-3}), the initial (i.e., at short correlation delay times τ) decrease of the ''nonlinear'' autocorrelation function is much faster than that of the linear one. Hence, our perturbation approach fails at short times. Indeed, the results of this section are based on the assumption that $C_{\Delta I}(\mathbf{r},0,\tau)$ and $C_{\Delta I}(\mathbf{r},\Delta\mathbf{r},\tau)$ in the nonlinear medium are close to their values in the linear one. Applying the factorization approximation, we find that this implies that $1-|g_1^{(NL)}(\tau)|^2\approx 1-|g_1^{(L)}(\tau)|^2$. Although this condition seems to hold well for sufficiently large τ, it can be violated at small τ, where, as follows from Fig. 1, $1-|g_1^{(NL)}(\tau)|^2$ can

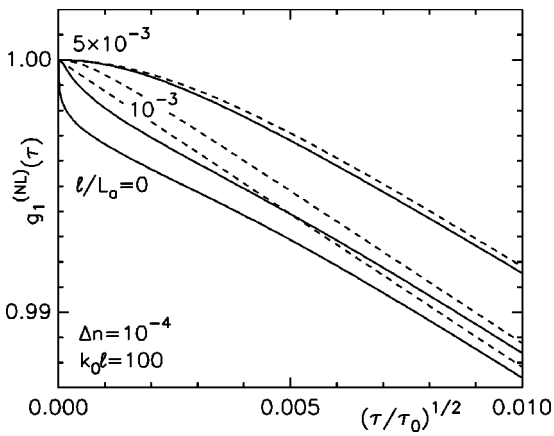

FIG. 1. Normalized temporal autocorrelation function of a wave diffusely reflected from a semi-infinite nonlinear disordered medium, calculated using perturbation theory for $\Delta n = 10^{-4}$, $k_0 l = 100$, and the values of l/L_a indicated near each curve (solid lines). Dashed lines show corresponding results for a linear medium ($\Delta n = 0$).

become much larger than $1-|g_1^{(L)}(\tau)|^2$, if absorption is sufficiently weak.

To estimate the region of validity of our perturbation approach, we require that the linear contribution to the dephasing $\langle \Delta\varphi^2(\tau)\rangle_s$ given by Eq. (5), should be considerably greater than the sum of nonlinear contributions [Eqs. (26)–(28)]. As the longest path length contributing to the integral of Eqs. (4) and (33) is $s \sim l/\alpha^2$, we obtain the conditions of validity of the perturbation theory in the form:

$$\Delta n^2 \alpha(\tau)^{-2}[k_0 l + \alpha(\tau)^{-1}] \ll 1. \tag{34}$$

Since $\alpha(\tau)$ is an increasing function of its argument, and $\alpha(0) = l/L_a$, condition $\Delta n^2 (L_a/l)^2 [k_0 l + L_a/l] \ll 1$ ensures Eq. (34) at any τ. It is the case for the upper curve of Fig. 1, corresponding to $l/L_a = 5\times 10^{-3}$. For such a strong absorption, the perturbation theory is valid at any τ, and the nonlinear autocorrelation function is close to the linear result. By contrast, if $\Delta n^2 (L_a/l)^2 [k_0 l + L_a/l] > 1$, the perturbation approach can be applied only for sufficiently long correlation times $\tau > \tau_c$, where the critical time τ_c is determined by Eq. (34). For the lower curve of Fig. 1, corresponding to $l/L_a = 0$, we find $(\tau_c/\tau_0)^{1/2} \approx 2\times 10^{-3}$. It is worthwhile to note that in the absence of absorption, even an infinitely small nonlinearity ($\Delta n \to 0$) suffices for the perturbation theory to fail at $\tau/\tau_0 \to 0$. Our condition of validity of the perturbation approach (34) is consistent with the result of Spivak and Zyuzin [70], who have shown that the perturbation analysis of the sensitivity of speckle pattern in a nonlinear disordered medium to changes of scattering potential fails for $\Delta n^2 (L/l)^3 > 1$, where $L \gg l$ is the typical size of the medium.

VI. SELF-CONSISTENT ANALYSIS

As demonstrated in the previous section, the perturbation theory fails to describe the temporal autocorrelation function of wave diffusely reflected from a nonlinear medium for $\tau < \tau_c$, where τ_c is defined by Eq. (34). To calculate $g_1^{(NL)}(\tau)$ at such short times, one has to use a nonperturbative, self-consistent analysis. However, the possibility of performing such an analysis is considerably limited by the mathematical complexity of the considered problem. To make the self-consistent analysis possible, we adopt the following two additional assumptions:

(iii) The statistics of a wave field scattered in a weakly nonlinear disordered medium, is close to Gaussian. Consequently, the factorization approximation holds in a weakly nonlinear medium: $C_{\delta I}(\mathbf{r},\tau) \simeq |C_\psi(\mathbf{r},\tau)|^2$.

(iv) The functional form of the $\mathbf{r}$ dependence of $g_1^{(NL)}(\mathbf{r},\tau)$ is the same as that of $g_1^{(L)}(\mathbf{r},\tau)$: $g_1^{(NL)}(\mathbf{r},\tau) = \exp[-\beta(\tau) z/l]$, where $\beta(\tau)$ is some unknown function, which can depend not only on τ but also on other parameters of the problem (namely, on l/L_a, $k_0 l$, Δn).

Strictly speaking, the above assumptions define a sort of perturbation theory, but now we do not limit the values of deviations of intensity and field correlation functions from their values in the linear case. Instead, we assume that the nonlinearity does not cause significant modifications of the *statistics* of scattered waves [assumption (iii)] and of the *functional form* of the field correlation function [assumption (iv)]. Note that now the autocorrelation function $g_1^{(NL)}(\mathbf{r},\tau)$ can deviate significantly from $g_1^{(L)}(\mathbf{r},\tau)$. Condition (iv) fixes the functional form of this deviation, but implies no constraints on its absolute value.

Obviously, both the assumptions (iii) and (iv) require the nonlinearity to be weak. Assumption (iii) is justified under the same conditions as (i) (see Sec. II), since the Gaussian statistics of the *total* scattered wave field $\psi(\mathbf{r},t)$ is a consequence of the complete randomization of phases of *partial* waves arriving at $\mathbf{r}$. The reason for the randomization is that the typical distance l between individual scattering events in a multiple-scattering sequence is much larger than the wavelength ($kl \gg 1$) [8]. Obviously, such a mechanism of phase randomization is equally effective in both linear and weakly nonlinear media, as long as (i) holds.

To justify the ansatz of assumption (iv), we apply Eq. (4) and write $g_1^{(NL)}$ as

$$g_1^{(NL)}(\mathbf{r},\tau) \approx \left(\frac{3z^2}{4\pi l}\right)^{1/2} \exp\left(\frac{z}{L_a}\right) \int_0^{s_1(\tau)} \frac{1}{s^{3/2}} \exp\left(-\frac{3z^2}{4ls}\right) ds, \tag{35}$$

where we assumed that $s/l_a + (1/2)\langle \Delta\varphi^2(\tau)\rangle_s$ increases monotonically with s and becomes of order unity at $s = s_1(\tau) \gg s_0 = z^2/(2l)$. After performing integration, Eq. (35) can be approximately rewritten as $\exp(z/L_a)\{1 - z[3/(\pi l s_1)]^{1/2}\} \simeq \exp[-\beta(\tau) z/l]$ with $\beta z/l \simeq z(l s_1)^{-1/2} - z/L_a \ll 1$. In the opposite limit of $\beta z/l \gg 1$, $g_1^{(NL)}(\mathbf{r},\tau)$ vanishes and the functional form of its z dependence is of no importance. Anyway, it will be seen from the following that the exact functional form of the $\mathbf{r}$ dependence of $g_1(\mathbf{r},\tau)$ is not of crucial importance, since g_1 is integrated over the whole medium during the calculation of the correlation function of diffusely reflected wave.

Making use of Eqs. (19)–(21) and relying on the assumptions (iii) and (iv), we recalculate the nonlinear contributions to the dephasing $\langle \Delta\varphi^2(\tau)\rangle_s$. Due to the assumption (iv), the

result can be obtained from Eqs. (26)–(28) by a simple substitution: $\alpha(\tau)\to\beta(\tau)+\alpha(0)$. Recalling that $\alpha(0)=l/L_a$, we obtain

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(1)}=2\Delta n\frac{\tau}{\tau_0}\left[1-H\left(\frac{l}{L_a}\sqrt{\frac{s}{12l}}\right)\right]\frac{s}{l}, \tag{36}$$

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(2)}=2\pi k_0 l\Delta n^2\left\{H\left(\left(\beta+\frac{l}{L_a}\right)\sqrt{\frac{s}{3l}}\right)-H\left(\frac{l}{L_a}\sqrt{\frac{s}{3l}}\right)\right\}\frac{s}{l}, \tag{37}$$

$$\langle\Delta\varphi^2(\tau)\rangle_s^{(3)}\simeq 6\Delta n^2\times\begin{cases}\beta(s/l)^2, & \beta\sqrt{s/l}\leqslant 1,\ (l/L_a)\sqrt{s/l}\leqslant 1,\\ (s/l)^{3/2}, & \beta\sqrt{s/l}>1,\ (l/L_a)\sqrt{s/l}\leqslant 1,\\ \beta(L_a/l)^3\sqrt{s/l}, & \beta\sqrt{s/l}\leqslant 1,\ (l/L_a)\sqrt{s/l}>1,\\ (L_a/l)^3, & \beta\sqrt{s/l}>1,\ (l/L_a)\sqrt{s/l}>1.\end{cases} \tag{38}$$

As follows from Eqs. (36)–(38), the nonlinear dephasing depends on the unknown function β. Since the temporal autocorrelation function of diffusely reflected wave $g_1^{(NL)}(\tau)=\exp[-\beta(\tau)]$ is determined by β as well, Eqs. (32) and (33) allow us to formulate a self-consistent equation for β:

$$\exp[-\beta(\tau)]=F(\beta(\tau)), \tag{39}$$

where $F(\beta)=W[\langle\Delta\varphi^2(\tau)\rangle_s]/W[0]$, $W[\ldots]$ is defined by Eq. (33), and $\langle\Delta\varphi^2(\tau)\rangle_s$ is a sum of terms given by Eqs. (5) and (36)–(38). Equation (39) is the main result of our self-consistent analysis. Although the functional form of $F(\beta)$ is rather complicated, and Eq. (39) cannot be solved analytically, the numerical solution is straightforward and can be carried out for any values of Δn, l/L_a, $k_0 l$, and τ. Equation (39) can be considered as a self-consistent equation for the autocorrelation function of diffusely reflected wave, since $\beta(\tau)$ and $g_1^{(NL)}(\tau)$ are directly related. It is worthwhile to note that in the absence of nonlinearity ($\Delta n=0$), Eq. (39) yields $\beta(\tau)=\alpha(\tau)-l/L_a$, and hence Eq. (8) is recovered Lor $g_1^{(L)}(\tau)$.

VII. RESULTS AND DISCUSSION

We start the analysis of Eq. (39) from the case of immobile scatterers, taking a limit of $\tau/\tau_0\to 0$. We denote the autocorrelation functions corresponding to this limit by $g_1^{(L),(NL)}(0^+)$ in order to distinguish them from $g_1^{(L),(NL)}(0)$, which correspond to $\tau=0$.[1] Obviously, $g_1^{(L)}(0^+)=g_1^{(L)}(0)=1$. In a nonlinear medium, Eqs. (37) and (38) can still contribute to the dephasing even for $\tau/\tau_0\to 0$. These contributions are insensitive to the sign of Δn. A corresponding value of $\beta(0^+)$ and, consequently, of the field autocorrelation function $g_1^{(NL)}(0^+)$ can be found by solving Eq. (39) numerically. In Fig. 2, we plot the left-hand and the right-hand sides of Eq. (39) for fixed $|\Delta n|=10^{-4}$, $k_0 l=100$, and for several values of l/L_a. If absorption is strong ($l/L_a\gtrsim 3\times 10^{-3}$ for considered $|\Delta n|$ and $k_0 l$), Eq. (39) has a unique solution $\beta(0^+)=0$, which corresponds to $g_1^{(NL)}(0^+)=1$. However, a second solution $\beta(0^+)>0$ appears for sufficiently weak absorption ($l/L_a<3\times 10^{-3}$). The point of appearance of the second solution is a *bifurcation* point of Eq. (39). To choose the solution realizable in a physical system, we note that the first solution $[\beta(0^+)=0]$ exists only for $\tau/\tau_0=0$, and disappears at finite τ/τ_0, since $F(0)<1$ for $\tau/\tau_0>0$. This solution is therefore inaccessible by continuity, and ''unstable'' with respect to small scatterer displacements. The physically realizable solution should represent the limit of $\beta(\tau)$ for $\tau/\tau_0\to 0$, which is given by the second solution of Eq. (39). It is therefore this solution that one expects to be realized in a real physical system.

The fact that Eq. (39) can have a positive solution

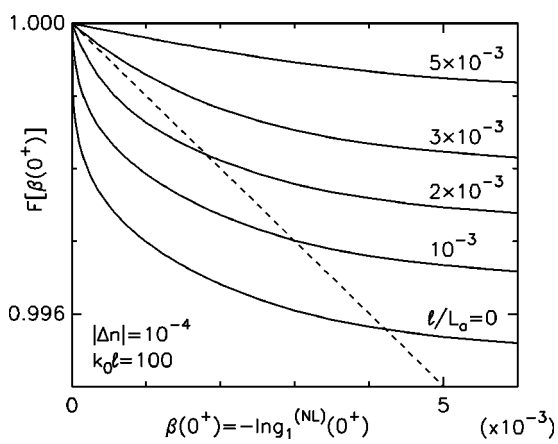

FIG. 2. Graphical solution of Eq. (39) at $\tau/\tau_0\to 0$. Solid lines show $F[\beta(0^+)]$ for $|\Delta n|=10^{-4}$, $k_0 l=100$, and the values of l/L_a indicated near each curve. Dashed line is $\exp[-\beta(0^+)]$. If absorption is weak ($l/L_a=0$, 10^{-3}, and 2×10^{-3}), Eq. (39) has two solutions $\beta(0^+)=0$ and $\beta(0^+)>0$, while for strong absorption ($l/L_a=3\times 10^{-3}$ and 5×10^{-3}), the second solution disappears.

[1]Although we take the limit of $\tau/\tau_0\to 0$, we assume $\tau\gg T_{jump}$ where $T_{jump}\gg 2\pi/\omega$ is the typical time required for the system to ''jump'' from one solution of Eq. (1) to another. The time autocorrelation function $g_1^{(NL)}(\tau)$ is independent of τ for $\tau\gg T_{jump}$ and $\tau/\tau_0\to 0$ [71].

$\beta(0^+)>0$ is a very important issue, since $\beta(0^+)>0$ leads to $g_1^{(NL)}(0^+)=\exp[-\beta(0^+)]<1$. A value of the temporal autocorrelation function, which is less than unity, is commonly associated with temporal fluctuations of scattered waves. In the considered case, however, the reason for these fluctuations is not the motion of scatterers, as the limit of $\tau/\tau_0 \to 0$ corresponds to immobile scatterers. The fluctuations are *spontaneous* and represent a clear signature of *instability* of the multiple-scattering speckle pattern.[2]

Despite a rather complicated structure of the function $F(\beta)$ in Eq. (39), a relation between the parameters of the problem corresponding to the onset of the speckle pattern instability can be found analytically. As follows from Fig. 2, the initial [at $\beta(0^+)=0$] decay of $F[\beta(0^+)]$ should be faster than the decay of $\exp[-\beta(0^+)]$, for the second solution of Eq. (39) to appear. A surface in a three-dimensional space of the problem parameters Δn, l/L_a, $k_0 l$, separating the stable $[\beta(0^+)=0]$ and unstable $[\beta(0^+)>0]$ regions, is therefore given by the equation

$$\frac{\partial}{\partial\beta(0^+)}F(\beta(0^+))|_{\beta(0^+)=0}=-1. \tag{40}$$

Recalling that $L_a/l \gg 1$, $k_0 l \gg 1$ is assumed, we obtain from Eq. (40):

$$p=\Delta n^2\left(\frac{L_a}{l}\right)^2\left[k_0 l+\frac{L_a}{l}\right]\simeq 1, \tag{41}$$

where we introduce a control parameter p, and a numerical factor of order unity is omitted. If $p<1$, the multiple-scattering speckle pattern is stable $[g_1^{(NL)}(0^+)=1]$, while for $p>1$, an instability shows up leading to $g_1^{(NL)}(0^+)<1$. A striking feature of Eq. (41) is that p can become larger than unity even for very small $|\Delta n|$, provided that the extensive parameter L_a/l is large enough. Our condition of the speckle pattern stability $p<1$ agrees with the condition of validity of the perturbation theory developed in Sec. V [Eq. (34)], evaluated at $\tau/\tau_0=0$. This readily explains the failure of the perturbation theory for short correlation times and weak absorption: perturbation approach is not suitable for description of unstable regimes. Moreover, the fact that the condition of validity of the perturbation theory and the speckle pattern stability condition agree indicates that the additional assumptions (iii) and (iv) of Sec. VI are not essential for obtaining unstable regimes.

To illustrate our self-consistent theoretical framework, we solve Eq. (39) numerically for $\tau/\tau_0 \to 0$, and plot the resulting temporal autocorrelation function of diffusely reflected wave, $g_1^{(NL)}(0^+)$, in Fig. 3. As discussed above, $g_1^{(NL)}(0^+)<1$ corresponds to spontaneous fluctuations of scattered waves, which is a manifestation of the speckle pattern instability. It follows from Fig. 3, that in accordance with Eq. (41), an infinitely small $|\Delta n|$ is sufficient to make the speckle pattern unstable in the absence of absorption, while a certain threshold degree of nonlinearity is required to destabilize the speckle pattern in a dissipative medium. In the absence of absorption, Eq. (39) always has two solutions: $\beta(0^+)=0$ and $\beta(0^+)>0$, corresponding to $g_1^{(NL)}(0^+)=1$ and $g_1^{(NL)}(0^+)<1$, respectively. As discussed above, it is the second solution, shown by a dashed line in Fig. 3, which is the physically realizable one. In contrast, if $l/L_a \neq 0$, $|\Delta n|$ should be greater than some threshold value for the second solution $\beta(0^+)>1$ to appear. Threshold values of $|\Delta n|$ following from Eq. (41) are shown in Fig. 3 by arrows. For a nonabsorbing, elastically scattering medium ($l/L_a=0$), the value of $g_1^{(NL)}(0^+)$ can be estimated analytically. Indeed, at $\tau/\tau_0 \to 0$ the principal contribution to $\langle\Delta\varphi^2(\tau)\rangle_s$ is given by $\langle\Delta\varphi^2(\tau)\rangle_s^{(2)}$ for $s/l \ll (k_0 l)^2$, and by $\langle\Delta\varphi^2(\tau)\rangle_s^{(3)}$ for $s/l \gg (k_0 l)^2$. This allows us to put $\langle\Delta\varphi^2(\tau)\rangle_s \approx \langle\Delta\varphi^2(\tau)\rangle_s^{(2)}$ if $s/l<(k_0 l)^2$, and $\langle\Delta\varphi^2(\tau)\rangle_s \approx \langle\Delta\varphi^2(\tau)\rangle_s^{(3)}$ if $s/l>(k_0 l)^2$. Integration in Eq. (33) can be then carried out, and Eq. (39) is easily solved, yielding

$$g_1^{(NL)}(0^+)\simeq 1-\begin{cases} 2|\Delta n|^{2/3}, & |\Delta n|<(k_0 l)^{-3/2}, \\ 3|\Delta n|\sqrt{k_0 l}, & |\Delta n|>(k_0 l)^{-3/2}. \end{cases} \tag{42}$$

This result agrees well with the numerical solution of Eq. (39) presented in Fig. 3.

The physical origin of the instability of speckle pattern for $p>1$ can be revealed by realizing that the system ''coherent wave + nonlinear disordered medium'' has a positive three-dimensional feedback. In a nonlinear medium, an infinitely

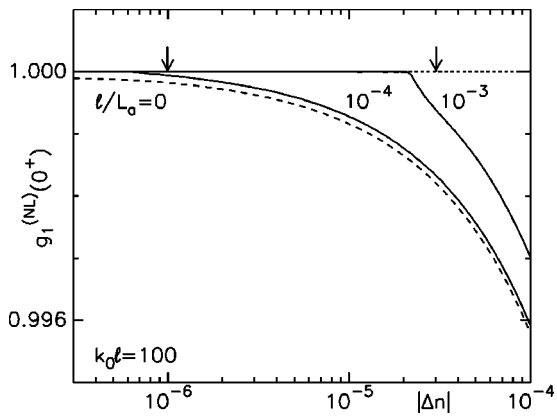

FIG. 3. ''Bifurcation diagram'' for a wave scattered in a semi-infinite nonlinear disordered medium. Solid lines show the temporal autocorrelation function of diffusely reflected wave at $\tau/\tau_0 \to 0$ (immobile scatterers) for $k_0 l=100$ and the values of l/L_a indicated near each curve (dashed line is for $l/L_a=0$). The threshold values of $|\Delta n|$ following from Eq. (41) are shown by arrows. $g_1^{(NL)}(0^+)<1$ corresponds to spontaneous fluctuations of scattered wave, which is a manifestation of the speckle pattern instability. Dotted line is the linear result $g_1^{(NL)}(0^+)=1$.

[2]Obviously, Eq. (1) cannot be used to study the dynamics of spontaneous fluctuations. Our analysis only allows the revealing of the presence of such fluctuations, and to calculate their time autocorrelation function $g_1^{(NL)}(\tau)$ for $2\pi/\omega \ll T_{jump} \ll \tau$.

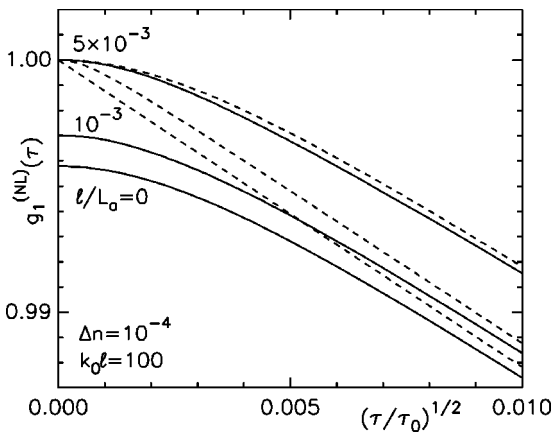

FIG. 4. The same as Fig. 1, but using a self-consistent approach instead of the perturbation theory. For the two lower curves, $g_1^{(NL)}(0^+)<1$ and the speckle pattern is unstable.

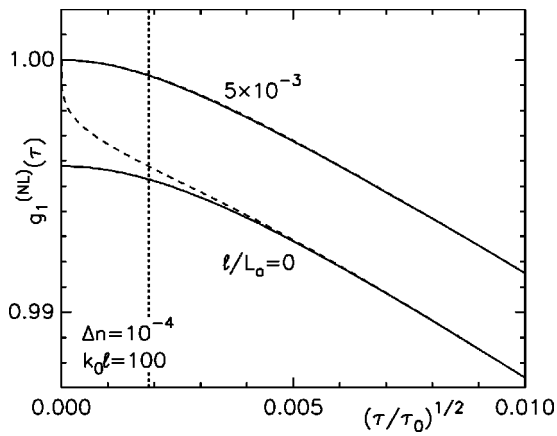

FIG. 5. Comparison of the normalized temporal autocorrelation functions calculated using a self-consistent (solid lines) and perturbation (dashed lines) theories. Dotted vertical line indicates the limit τ_c of validity of the perturbation theory for $l/L_a=0$. The perturbation theory is valid only for $\tau>\tau_c$, while for $\tau<\tau_c$ the perturbation result deviates significantly from the self-consistent solution. For sufficiently strong absorption ($l/L_a=5\times10^{-3}$), the perturbation theory holds at any τ: the perturbation and the self-consistent results are indistinguishable.

small perturbation of the wave intensity $I(\mathbf{r},t)$ produces a change of the local refractive index, which alters the phases of waves propagating in the medium and, consequently, affects their mutual interference. Since it is this interference that determines $I(\mathbf{r},t)$, the loop of the feedback is closed. For $p\gtrsim1$, the feedback is sufficiently strong to compensate for the (diffusive on average) spreading of the initial intensity perturbation, and the speckle pattern $I(\mathbf{r},t)$ is unstable. It is worthwhile to note that unstable regimes are not exceptional in nonlinear wave systems and, in particular, in optical systems (see, e.g., Refs. [72–76]).

Our Eq. (39) is in no way limited to the case of $\tau/\tau_0\to0$, and can be used to compute the temporal autocorrelation function of diffusely reflected wave at any $\tau/\tau_0>0$. In the latter case, one should take into account all the four dephasing terms given by Eqs. (5) and (36)–(38). The results of the numerical solution of Eq. (39) are shown in Fig. 4. For weak absorption ($l/L_a=0,10^{-3}$), $p>1$ and the speckle pattern is unstable [$g_1^{(NL)}(0^+)<1$]. As τ/τ_0 increases, the difference between the ''nonlinear'' and ''linear'' curves becomes less pronounced. For strong absorption ($l/L_a=5\times10^{-3}$), p becomes less than unity and stability of the speckle pattern is recovered [$g_1^{(NL)}(0^+)=1$]. We remind that the temporal autocorrelation function $g_1^{(L)}(\tau)$, corresponding to a linear medium, always equals 1 for $\tau/\tau_0=0$, as shown by dashed lines in Fig. 4.

It is instructive to compare the results obtained using the perturbation theory of Sec. V and the self-consistent approach of Sec. VI. Such a comparison is shown in Fig. 5. The two upper curves, corresponding to relatively strong absorption ($l/L_a=5\times10^{-3},p<1$), are almost indistinguishable, which means that for $p<1$, the perturbation theory works very well. In contrast, for $p>1$ (see the two lower curves of Fig. 5, corresponding to $l/L_a=0$), the perturbation and the self-consistent curves are close only at the right from the dotted vertical line, showing the minimum time τ_c at which the perturbation theory is valid [see Eq. (34)]. For $\tau\lesssim\tau_c$, the perturbation and the self-consistent results disagree significantly, which confirms our conclusion about the failure of the perturbation approach at short correlation delay times.

VIII. CONCLUSION

We are now in a position to answer the two central questions formulated in the introductory section:

(a) The phenomenon of multiple scattering is capable of providing a positive feedback for a coherent wave propagating in a nonlinear disordered medium.

(b) The onset of the speckle pattern instability occurs when the control parameter

$$p=\Delta n^2\left(\frac{L_a}{l}\right)^2\left[k_0l+\frac{L_a}{l}\right] \tag{43}$$

becomes of order or larger than unity. The speckle pattern is stable for $p<1$.

The instability of the multiple-scattering speckle pattern manifests itself in spontaneous fluctuations of the scattered wave field and intensity. The following features are characteristic for the development of the instability. First, the development of the instability is independent of the sign of nonlinearity. This is not common for nonlinear waves since the instability is often due to self-focusing phenomena, which only occur for $n_2>0$ [72–76]. The instability of waves in a *disordered* nonlinear medium has nothing to do with the self-focusing, and occurs at relatively weak nonlinearities, when the self-focusing can be neglected. Second, in the absence of absorption, the speckle pattern is unstable for any (even infinitely small) value of the nonlinearity strength $|\Delta n|$, while in a dissipative medium $|\Delta n|$ should exceed a certain threshold value for the instability to show up. Finally, the instability results in a value of the autocorrelation func-

tion $g_1^{(NL)}(\tau)$ of scattered wave, which is smaller than 1 for $\tau/\tau_0 \to 0$ (i.e., in the absence of scatterer motion). This is a clear signature of spontaneous fluctuations of the multiple-scattered speckle pattern, and should be observable in experiments.

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APPENDIX A: SPATIOTEMPORAL FIELD CORRELATION IN A LINEAR MEDIUM

In this appendix, we provide a derivation of the spatiotemporal correlation function, $C_\psi(\mathbf{r},\Delta\mathbf{r},\tau)=\langle \psi(\mathbf{r}-\Delta\mathbf{r}/2,t)\psi^*(\mathbf{r}+\Delta\mathbf{r}/2,t+\tau)\rangle$, of a random field $\psi(\mathbf{r},t)$ in the bulk of disordered medium. Starting from the linear wave equation [Eq. (1) with $\varepsilon_2=0$], we obtain the Bethe-Salpeter equation in the form [11,80,8]:

$$\begin{aligned} C_\psi(\mathbf{r},\Delta\mathbf{r},\tau) &= \langle \psi(\mathbf{r}-\Delta\mathbf{r}/2,t)\rangle\langle \psi^*(\mathbf{r}+\Delta\mathbf{r}/2,t)\rangle \\ &\quad + \int d\mathbf{r}_a \int d\mathbf{r}_b \int d\mathbf{r}_c \int d\mathbf{r}_d \overline{G}(\mathbf{r}-\Delta\mathbf{r}/2,\mathbf{r}_a) \\ &\quad \times \overline{G}^*(\mathbf{r}+\Delta\mathbf{r}/2,\mathbf{r}_b) U(\mathbf{r}_a,\mathbf{r}_b,\mathbf{r}_c,\mathbf{r}_d) \\ &\quad \times C_\psi[(\mathbf{r}_c+\mathbf{r}_d)/2,\mathbf{r}_d-\mathbf{r}_c,\tau], \end{aligned} \tag{A1}$$

where the integrations are over the volume of disordered medium, $\overline{G}$ is the average Green function of the linear wave equation, and U is the irreducible four-point vertex. Far enough from the medium boundaries, we can replace $\overline{G}$ by its value in the infinite medium:

$$\overline{G}(\mathbf{r}_1,\mathbf{r}_2) = -\frac{1}{4\pi|\mathbf{r}_1-\mathbf{r}_2|}\exp\left[\left(ik-\frac{1}{2l}\right)|\mathbf{r}_1-\mathbf{r}_2|\right]. \tag{A2}$$

Now we assume that point scatterers in the medium undergo Brownian motion, and that the correlation function of the dielectric function fluctuations is given by Eq. (3). For $\tau \ll \tau_0$, we can neglect the τ dependence of U in Eq. (A1), which in the limit of $kl \gg 1$ becomes

$$U(\mathbf{r}_a,\mathbf{r}_b,\mathbf{r}_c,\mathbf{r}_d) = \frac{4\pi}{l}\delta(\mathbf{r}_a-\mathbf{r}_c)\delta(\mathbf{r}_b-\mathbf{r}_d)\delta(\mathbf{r}_a-\mathbf{r}_b). \tag{A3}$$

Equation (A1) then reduces to

$$\begin{aligned} C_\psi(\mathbf{r},\Delta\mathbf{r},\tau) &= \frac{4\pi}{l}\int d\mathbf{r}_a \overline{G}(\mathbf{r}-\Delta\mathbf{r}/2,\mathbf{r}_a) \\ &\quad \times \overline{G}^*(\mathbf{r}+\Delta\mathbf{r}/2,\mathbf{r}_a) C_\psi(\mathbf{r}_a,0,\tau), \end{aligned} \tag{A4}$$

where we assumed the coherent field $\langle \psi(\mathbf{r},t)\rangle$ to be negligible.

Now we remind that $C_\psi(\mathbf{r}_a,0,\tau)\equiv C_\psi(\mathbf{r}_a,\tau)$ is a slow-varying function of $\mathbf{r}_a$. We can therefore pull it out of the integral in Eq. (A4), taking its value at $\mathbf{r}$. Performing the remaining integral, we find

$$C_\psi(\mathbf{r},\Delta\mathbf{r},\tau) = \frac{\sin(k\Delta r)}{k\Delta r}\exp\left(-\frac{\Delta r}{2l}\right) C_\psi(\mathbf{r},\tau). \tag{A5}$$

This equation relates the spatiotemporal correlation of the field with its purely temporal correlation. Equation (A5) holds for any sample geometry and source distribution, far enough from boundaries and sources, and for $\tau \ll \tau_0$. In the case of a plane wave incident at the surface $z=0$ of a semi-infinite disordered medium, occupying the $z>0$ half-space,

$$C_\psi(\mathbf{r},\tau) = I_0 \exp[-\alpha(\tau)(z/l)] \tag{A6}$$

with $\alpha^2(\tau)=3\tau/(2\tau_0)+(l/L_a)^2$, leading to Eq. (9) in the main text. In the factorization approximation (10), the square of Eq. (A5) gives the short-range correlation function of intensity fluctuations.

APPENDIX B: LONG-RANGE SPATIOTEMPORAL CORRELATION OF INTENSITY FLUCTUATIONS IN A LINEAR MEDIUM

To calculate the spatiotemporal correlation function of intensity fluctuations, $C_{\delta I}(\mathbf{r},\Delta\mathbf{r},\tau)=\langle \delta I(\mathbf{r}-\Delta\mathbf{r}/2,t)\delta I(\mathbf{r}+\Delta\mathbf{r}/2,t+\tau)\rangle$, for $\Delta r > l$ we generalize the Langevin approach [9,11,90], which has been initially developed for calculation of purely spatial correlations. We assume that the spatiotemporal correlation function of Langevin random sources is determined by the short-range correlation function of intensity fluctuations. In the factorization approximation, the latter is given by the square of the field-field correlation function [Eq. (A5)]. Hence, the generalized Langevin equations read

$$D_p[\nabla^2 - 1/L_a^2]\delta I(\mathbf{r},t) = \mathrm{div}\mathbf{j}_{ext}(\mathbf{r},t), \tag{B1}$$

$$\begin{aligned} &\langle j_{ext}^{(i)}(\mathbf{r}-\Delta\mathbf{r}/2,t) j_{ext}^{(m)}(\mathbf{r}+\Delta\mathbf{r}/2,t+\tau)\rangle \\ &\quad = \frac{1}{3}\delta_{im}\frac{c^2 2\pi l}{k^2}|C_\psi(\mathbf{r},\tau)|^2\delta(\Delta\mathbf{r}), \end{aligned} \tag{B2}$$

where $D_p = cl/3$ is the diffusion coefficient, and c is the speed of wave in the medium. In the case of a plane wave incident upon a boundary $z=0$ of a semi-infinite disordered medium, it is convenient to make a two-dimensional Fourier transform of Eq. (B1) in the $\{x,y\}$ plane. Then the equations corresponding to $\delta I(\mathbf{K}_1,z_1,t_1)$ and $\delta I(\mathbf{K}_2,z_2,t_2)$ are multiplied, and the product is ensemble averaged. Further transformations, which are equivalent to those discussed in Ref. [11], yield

$$\langle \delta I(\mathbf{K}_1,z_1,t_1)\delta I(\mathbf{K}_2,z_2,t_2)\rangle = I_0^2\frac{6\pi}{k^2 l}\delta(\mathbf{K}_1-\mathbf{K}_2)\int_0^\infty dz'\left[(\mathbf{K}_1\cdot\mathbf{K}_2)G(p,z_1,z')G(p,z_2,z') + \frac{\partial}{\partial z'}G(p,z_1,z')\frac{\partial}{\partial z'}G(p,z_2,z')\right]\exp\left[-2\alpha(\tau)\frac{z'}{l}\right], \tag{B3}$$

where $p^2=\mathbf{K}^2+1/L_a^2$, and

$$G(p,z_1,z') = -\frac{1}{p}\sinh(p\min\{z_1,z'\})\exp(-p\max\{z_1,z'\}) \tag{B4}$$

is the Green function of Eq. (B1). Evaluating Eq. (B3) and transforming the result back to the real space, after lengthy but straightforward algebra, we obtain

$$C_{\delta I}(\mathbf{r},\Delta\mathbf{r},\tau) = \frac{3}{(kl)^2}I_0^2\int_0^\infty dK\,K\,Q\left(K,\sqrt{K^2+l^2/L_a^2},\frac{z}{l},\frac{\Delta z}{l},\alpha(\tau)\right)J_0\left(K\frac{\Delta R}{l}\right), \tag{B5}$$

where

$$\begin{aligned} Q(K,p,\zeta,\Delta\zeta,\alpha) = {} & \exp(2p\zeta)\times\left[\frac{2\alpha^2+K^2-p^2}{4\alpha^3-4\alpha p^2} - \frac{(\alpha^2-p^2)(-K^2+p^2)+\alpha^2(K^2+p^2)\cosh(2p\zeta_1)+\alpha p(K^2+p^2)\sinh(2p\zeta_1)}{4\alpha\exp(2\alpha\zeta_1)(\alpha^2-p^2)p^2}\right] \\ & +\left(1+\frac{K^2}{p^2}\right)\frac{\sinh(p\zeta_1)\sinh(p\zeta_2)}{2\exp[2(\alpha+p)\zeta_2](\alpha+p)} + \frac{\exp(-p\zeta_2)\sinh(p\zeta_1)}{4\alpha p^2(\alpha+p)} \\ & \times\{p(p^2+2\alpha p-K^2)\{\cosh(p\zeta_2)\exp[-(2\alpha+p)\zeta_2]-\cosh(p\zeta_1)\exp[-(2\alpha+p)\zeta_1]\} \\ & +(p^3-2\alpha K^2-pK^2)\{\sinh(p\zeta_2)\exp[-(2\alpha+p)\zeta_2]-\sinh(p\zeta_1)\exp[-(2\alpha+p)\zeta_1]\}\}, \end{aligned} \tag{B6}$$

where $\zeta_1=\zeta-\Delta\zeta/2$ and $\zeta_2=\zeta+\Delta\zeta/2$.

Equations (B5) and (B6) cannot be evaluated in a general form. In the limits of $\alpha(\tau)\zeta,\alpha(\tau)\Delta R/l\ll 1$ we can, however, approximately replace Q in Eq. (B5) by

$$Q(K,p,\zeta,\Delta\zeta,\alpha)\approx\frac{1}{2K}[\exp(-K\Delta\zeta)-\exp(-2K\zeta)] \times\exp[-2\alpha(\tau)\zeta], \tag{B7}$$

which yields Eq. (15) in the main text.

APPENDIX C: DENSITY OF WAVE PATHS IN A DISORDERED MEDIUM

Propagation of waves in a disordered medium can be interpreted in terms of partial waves traveling along various paths inside the medium. The spatial distribution of such paths and their relative weights depend on the scattering properties of the medium, and on the geometry of the sample. In the case of multiple scattering, the simplest and, at the same time, sufficiently accurate model of wave propagation is the *diffusion* model. According to this model, wave paths in the medium coincide with trajectories of Brownian particles. The probability $G(\mathbf{r}_1,\mathbf{r}_2,s)$ for a path of length s to pass from $\mathbf{r}_1$ to $\mathbf{r}_2$ is then given by a solution of the diffusion equation, which in the absence of absorption reads [86,89]:

$$\frac{d}{ds}G(\mathbf{r}_1,\mathbf{r}_2,s)-\frac{1}{3}l\nabla^2G(\mathbf{r}_1,\mathbf{r}_2,s)=\delta(\mathbf{r}_1-\mathbf{r}_2)\delta(s), \tag{C1}$$

where l is the mean-free-path. Commonly used boundary conditions for Eq. (C1) consist in putting $G=0$ at open boundaries and $\nabla_n G=0$ at reflecting boundaries of the sample (where ∇_n denotes the normal derivative of G). $G(\mathbf{r}_1,\mathbf{r}_2,s)$ is called the Green function, or the propagator. For a semi-infinite medium occupying the half-space $z>0$, one finds

$$G(\mathbf{r}_1,\mathbf{r}_2,s)=\left(\frac{3}{4\pi ls}\right)^{3/2}\left\{\exp\left[-\frac{3}{4ls}(\Delta R^2+\Delta z^2)\right] - \exp\left[-\frac{3}{4ls}(\Delta R^2+Z^2)\right]\right\}, \tag{C2}$$

where cylindrical coordinates are used: $\mathbf{r}_i=\{\mathbf{R}_i,z_i\}$, $\Delta\mathbf{R}=\mathbf{R}_1-\mathbf{R}_2$, $\Delta z=z_1-z_2$, and $Z=z_1+z_2$.

Following Ref. [86], we introduce $\rho_s(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$, the *density distribution* of paths of length s, as a number of visits of a given site $\mathbf{r}_2$ inside $d^3\mathbf{r}_2$ in the ensemble of paths of length s starting at $\mathbf{r}_1$ and ending at $\mathbf{r}_3$, over the total length of the ensemble distinct paths:

$$\rho_s(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)=\frac{1}{sG(\mathbf{r}_1,\mathbf{r}_3,s)}\times\int_0^s dpG(\mathbf{r}_1,\mathbf{r}_2,p)G(\mathbf{r}_2,\mathbf{r}_3,s-p). \tag{C3}$$

$\rho_s(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$ describes the probability density for a path of a given length s, starting at $\mathbf{r}_1$ and ending at $\mathbf{r}_3$, to pass through $\mathbf{r}_2$. This quantity is normalized:

$$\int d^3\mathbf{r}_2\rho_s(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)=1, \tag{C4}$$

where the integration is performed over the volume of disordered medium.

As the Green function G is known [Eq. (C2)], the calculation of $\rho_s(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$ is straightforward. For diffusely reflected paths, assuming that the first and the last scattering events take place at $z=l$, we obtain

$$\rho_s(l,\mathbf{r},l)\equiv\rho_s(\mathbf{r})=\frac{1}{A}\frac{6z}{ls}\exp\left(-\frac{3z^2}{ls}\right), \tag{C5}$$

where $A\to\infty$ is the surface of the semi-infinite medium. Equation (C5) defines the probability density for a diffusely reflected path of length s to pass through a vicinity of some point $\mathbf{r}=\{x,y,z\}$.

Generalizing definition of ρ_s, we define the probability density for a path of length s starting at $\mathbf{r}_1$ and ending at $\mathbf{r}_4$ to pass consequently through $\mathbf{r}_2$ and $\mathbf{r}_3$:

$$\rho_s(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)=\frac{2}{s^2G(\mathbf{r}_1,\mathbf{r}_4,s)}\int_0^s dp\times\int_0^{s-p}dqG(\mathbf{r}_1,\mathbf{r}_2,p)G(\mathbf{r}_2,\mathbf{r}_3,q)\times G(\mathbf{r}_3,\mathbf{r}_4,s-p-q). \tag{C6}$$

The normalization of Eq. (C6) is

$$\int d^3\mathbf{r}_2\int d^3\mathbf{r}_3\rho_s(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)=1. \tag{C7}$$

For a semi-infinite medium, we get

$$\rho_s(l,\mathbf{r},\mathbf{r}',l)\equiv\rho_s(\mathbf{r},\mathbf{r}')=\frac{1}{A^2}\frac{9}{2\pi l^2s^2}\times\left\{\frac{Z+\sqrt{\Delta R^2+\Delta z^2}}{\sqrt{\Delta R^2+\Delta z^2}}\times\exp\left[-\frac{3}{4ls}(Z+\sqrt{\Delta R^2+\Delta z^2})^2\right]-\frac{Z+\sqrt{\Delta R^2+Z^2}}{\sqrt{\Delta R^2+Z^2}}\times\exp\left[-\frac{3}{4ls}(Z+\sqrt{\Delta R^2+Z^2})^2\right]\right\}, \tag{C8}$$

where $\mathbf{r}=\{\mathbf{R},z\},\Delta\mathbf{R}=\mathbf{R}-\mathbf{R}',\Delta z=z-z',Z=z+z'$.

Although Eqs. (C5) and (C8) have been found for a nonabsorbing medium, it is easy to show that these results hold in the presence of *spatially-homogeneous* absorption as well. This stems from the fact that the attenuation of wave in a homogeneously absorbing medium depends only on the path *length*, while being independent of the path *geometry*. As a consequence, the Green function [Eq. (C2)] should be multiplied by a factor $\exp(-s/l_a)$, where l_a is the absorption length. This factor, however, disappears after the substitution of the Green function (C2) in Eqs. (C3) and (C6). Consequently, $\rho_s(\mathbf{r})$ and $\rho_s(\mathbf{r},\mathbf{r}')$ are independent of l_a and remain unchanged.

[1] P. Sheng, *Introduction to Wave Scattering, Localization, and Mesoscopic Phenomena* (Academic, Boston, 1995).
[2] *Wave Propagation in Complex Media*, edited by G. Papanicolaou (Springer-Verlag, Heidelberg, 1997).
[3] *New Aspects of Electromagnetic and Acoustic Wave Propagation*, edited by GDR POAN (Springer-Verlag, Heidelberg, 1998).
[4] *Wave Diffusion in Complex Media*, edited by J.-P. Fouque (Kluwer, Dordrecht, 1999).
[5] M.C.W. van Rossum and Th.M. Nieuwenhuizen, Rev. Mod. Phys. **71**, 313 (1999).
[6] J.W. Goodman, J. Opt. Soc. Am. **66**, 1145 (1976).
[7] A. Ishimaru, *Wave Propagation and Scattering in Random Media* (Academic, New York, 1978).
[8] B. Shapiro, Phys. Rev. Lett. **57**, 2168 (1986).
[9] A.Yu. Zyuzin and B.Z. Spivak, Zh. Éksp. Teor. Fiz. **93,** 994 (1987) [Sov. Phys. JETP **66**, 560 (1987)].
[10] M.J. Stephen and G. Cwilich, Phys. Rev. Lett. **59**, 285 (1987).
[11] R. Pnini and B. Shapiro, Phys. Rev. B **39**, 6986 (1989); Phys. Lett. A **157**, 265 (1991).
[12] R. Berkovits and S. Feng, Phys. Rep. **238**, 135 (1994).
[13] B. Shapiro, Phys. Rev. Lett. **83**, 4733 (1999).
[14] S.E. Skipetrov and R. Maynard, Phys. Rev. B **62**, 886 (2000).
[15] P.A. Lee and A.D. Stone, Phys. Rev. Lett. **55**, 1622 (1985).
[16] R. Berkovits, Phys. Rev. B **43**, 8638 (1991).
[17] B. Simons and B. Altshuler, Phys. Rev. Lett. **70**, 4063 (1993).
[18] A.A. Golubentsev, Zh. Éksp. Teor. Fiz. **86,** 47 (1984) [Sov. Phys. JETP **59**, 26 (1984)].
[19] G. Maret and P.E. Wolf, Z. Phys. B: Condens. Matter **65**, 409 (1987).
[20] M.J. Stephen, Phys. Rev. B **37**, 1 (1988).
[21] F. Scheffold, W. Härtl, G. Maret, and E. Matijević, Phys. Rev. B **56**, 10 942 (1997).

[22] F. Scheffold and G. Maret, Phys. Rev. Lett. **81**, 5800 (1998).
[23] A.A. Burkov and A.Yu. Zyuzin, Phys. Rev. B **55**, 5736 (1997).
[24] S.E. Skipetrov, S.S. Chesnokov, S.D. Zakharov, M.A. Kazaryan, and V.A. Shcheglov, Pis'ma Zh. Éksp. Teor. Fiz. **67,** 611 (1998) [JETP Lett. **67**, 635 (1998)].
[25] S.E. Skipetrov, S.S. Chesnokov, S.D. Zakharov, M.A. Kazaryan, N.P. Korotkov, and V.A. Shcheglov, Kvant. Elektron. (Moscow) **25,** 447 (1998) [Quantum Electron. **28**, 434 (1998)].
[26] D.J. Pine, D.A. Weitz, P.M. Chaikin, and E. Herbolzheimer, Phys. Rev. Lett. **60**, 1134 (1988).
[27] D.J. Pine, D.A. Weitz, G. Maret, P.E. Wolf, E. Herbolzheimer, and P.M. Chaikin, in *Scattering and Localization of Classical Waves in Random Media*, edited by P. Sheng (World Scientific, London, 1989), p. 312.
[28] D.J. Pine, D.A. Weitz, J.X. Zhu, and E. Herbolzheimer, J. Phys. (France) **51**, 2101 (1990).
[29] D.A. Weitz and D.J. Pine, in *Dynamic Light Scattering*, edited by W. Brown (Oxford University Press, New York, 1993), pp. 652–720.
[30] G. Maret, Curr. Opin. Colloid Interface Sci. **2**, 251 (1997).
[31] D.J. Durian and D.A. Weitz, Science **252**, 686 (1991).
[32] J.C. Earnshow and A.H. Jaafar, Phys. Rev. E **49**, 5408 (1994).
[33] A.D. Gopal and D.J. Durian, Phys. Rev. Lett. **75**, 2610 (1995).
[34] R. Höhler, S. Cohen-Addad, and H. Hoballah, Phys. Rev. Lett. **79**, 1154 (1997).
[35] A.D. Gopal and D.J. Durian, J. Opt. Soc. Am. A **14**, 150 (1997).
[36] H. Gang, A.H. Krall, and D.A. Weitz, Phys. Rev. E **52**, 6289 (1995).
[37] P. Hebraud, F. Lequeux, J.P. Munch, and D.J. Pine, Phys. Rev. Lett. **78**, 4657 (1997).
[38] V. Lisy and B. Brutovsky, Phys. Rev. E **59**, 3756 (1999) (comment on Ref. [36]).
[39] N. Menon and D.J. Durian, Phys. Rev. Lett. **79**, 3407 (1997).
[40] N. Menon and D.J. Durian, Science **275**, 1920 (1997).
[41] D.J. Durian, J. Phys. C **12**, A507 (2000).
[42] D.A. Boas, G. Nishimura, and A.G. Yodh, Proc. SPIE **2979**, 468 (1997).
[43] A. Palmer, T.G. Mason, X. Jingyuan, S.C. Kuo, and D. Wirtz, Biophys. J. **76**, 1063 (1999).
[44] R. Lohwasser and G. Soelkner, Appl. Opt. **38**, 2128 (1999).
[45] D.A. Boas, L.E. Campbell, and A.G. Yodh, Phys. Rev. Lett. **75**, 1855 (1995).
[46] D.A. Boas and A.G. Yodh, J. Opt. Soc. Am. A **14**, 192 (1997).
[47] M. Heckmeier, S.E. Skipetrov, G. Maret, and R. Maynard, J. Opt. Soc. Am. A **14**, 185 (1997).
[48] S.E. Skipetrov and I.V. Meglinskiĭ, Zh. Éksp. Teor. Fiz. **113,** 1213 (1998) [JETP **86**, 661 (1998)].
[49] M. Heckmeier and G. Maret, Opt. Commun. **148**, 1 (1998).
[50] H. Stark and T. Lubensky, Phys. Rev. Lett. **77**, 2229 (1996).
[51] H. Stark, M.H. Kao, K.A. Jester, T.C. Lubensky, A.G. Yodh, and P.J. Collings, J. Opt. Soc. Am. A **14**, 156 (1997).
[52] H. Stark and T.C. Lubensky, Phys. Rev. E **55**, 514 (1997).
[53] G. Nisato, P. Hébraud, J.-P. Munch, and S.J. Candau, Phys. Rev. E **61**, 2879 (2000).
[54] F. Scheffold, S. E. Skipetrov, S. Romer, and P. Schurtenberger (unpublished).
[55] V.M. Agranovich and V.E. Kravtsov, Phys. Rev. B **43**, 13 691 (1991).
[56] V.M. Agranovich and K.I. Grigorishin, Nonlinear Opt. **5**, 3 (1993).
[57] A. Heiderich, R. Maynard, and B.A. van Tiggelen, Opt. Commun. **115**, 392 (1995).
[58] V.M. Agranovich and V.E. Kravtsov, Phys. Lett. A **131**, 378 (1988); **131**, 387 (1988).
[59] V.M. Agranovich and V.E. Kravtsov, Zh. Éksp. Teor. Fiz. **95,** 484 (1989) [Sov. Phys. JETP **68**, 272 (1989)].
[60] V.E. Kravtsov, V.M. Agranovich, and K.I. Grigorishin, Phys. Rev. B **44**, 4931 (1991).
[61] K.M. Yoo, S. Lee, Y. Takiguchi, and R.R. Alfano, Opt. Lett. **14**, 800 (1989).
[62] V.E. Kravtsov, V.I. Yudson, and V.M. Agranovich, Solid State Commun. **72**, 1105 (1989).
[63] V.E. Kravtsov, V.I. Yudson, and V.M. Agranovich, Phys. Rev. B **41**, 2794 (1990).
[64] V.I. Yudson and P. Reineker, Physica B **175**, 71 (1991).
[65] J.C.J. Paasschens, M.J.M. de Jong, P.W. Brouwer, and C.W.J. Beenakker, Phys. Rev. A **56**, 4216 (1997).
[66] J.F. de Boer, A. Lagendijk, R. Sprik, and S. Feng, Phys. Rev. Lett. **71**, 3947 (1993).
[67] R. Bressoux and R. Maynard, Europhys. Lett. **50**, 460 (2000).
[68] A.J. van Wonderen, J. Phys. I **2**, 545 (1992).
[69] A.J. van Wonderen, Phys. Rev. B **50**, 2921 (1994).
[70] B. Spivak and A. Zyuzin, Phys. Rev. Lett. **84**, 1970 (2000).
[71] S.E. Skipetrov and R. Maynard, Phys. Rev. Lett. **85**, 736 (2000).
[72] H.M. Gibbs, *Optical Bistability: Controlling Light with Light* (Academic, New York, 1985).
[73] *Self-Organization in Optical Systems and Applications in Information Technology*, edited by M.A. Vorontsov and W.B. Miller (Springer-Verlag, Berlin 1995).
[74] G. D'Alessandro and W.J. Firth, Phys. Rev. Lett. **66**, 2597 (1991); Phys. Rev. A **46**, 537 (1992).
[75] F.T. Arecchi, S. Boccaletti, and P.L. Ramazza, Phys. Rep. **318**, 1 (1999).
[76] P.L. Ramazza, S. Residori, E. Pamapaloni, and A.V. Larichev, Phys. Rev. A **53**, 400 (1996).
[77] Y.R. Shen, *Principles of Nonlinear Optics* (Wiley, New York, 1984).
[78] J.V. Moloney and A.C. Newell, *Nonlinear Optics* (Addison-Wesley, Redwood City, CA, 1991).
[79] For the sake of concreteness, we use the ''optical'' language throughout the paper. Our results are, however, general, and apply to all types of scalar waves (in the case of optical waves, polarization effects are neglected).
[80] U. Frisch, in *Probabilistic Methods in Applied Mathematics*, edited by A.T. Bharucha Reid (Academic, New York, 1968).
[81] S.M. Rytov, Yu.A. Kravtsov, and V.I. Tatarskii, *Principles of Statistical Radiophysics* (Springer-Verlag, New York, 1989).
[82] R. Dashen, J. Math. Phys. **20**, 894 (1979).
[83] E.P. Gross, J. Math. Phys. **24**, 399 (1983).
[84] R.P. Feynman and A.R. Hibbs, *Quantum Mechanics and Path Integrals* (McGraw-Hill, New York, 1965).
[85] L.S. Schulman, *Techniques and Applications of Path Integration* (Wiley, New York, 1981).
[86] D. Bicout and R. Maynard, Physica A **199**, 387 (1993).
[87] This is called the Siegert relation, see, e.g., B.J. Berne and R.

Pecora, *Dynamic Light Scattering* (Wiley, New York, 1976), p. 40.

[88] A.A. Karabutov, I.M. Pelivanov, N.B. Podymova, and S.E. Skipetrov, Pis'ma Zh. Éksp. Teor. Fiz. **70,** 187 (1999) [JETP Lett. **70**, 183 (1999)].

[89] E. Akkermans, P.E. Wolf, R. Maynard, and G. Maret, J. Phys. (Paris) **49**, 77 (1988).

[90] One can check, however, that exactly the same result follows from a diagrammatic calculation where interaction of four ladders is described with help of the Hikami vertex.