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Symmetry between absorption and amplification in disordered media

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We address the issue of whether amplification, like absorption, suppresses wave transmission at large gain, as has been claimed in previous studies of wave propagation in active random media. A closer examination reveals that the paradoxical symmetry between absorption and amplification is an artifact of unphysical solutions from the time-independent wave equation. Solutions from the time-dependent equation demonstrate clearly that when gain is above the threshold, the amplitude of both the transmitted and the reflected wave actually increases with time, apparently without bound. The implications of the current finding is discussed. [S0163-1829(99)50314-7]

Recent observations[1] of laserlike emission from dye solution immersed with TiO_2 nanoparticles have stimulated intensive theoretical efforts[2–13] to investigate the properties of disordered media which are optically active. With enhanced optical paths from multiple scattering, random systems are expected to possess a reduced gain threshold for lasing.[14,15] Correspondingly, one would expect the transmission in disordered systems to be enhanced with gain. Surprisingly, transfer matrix calculations[3,4] based on time-independent wave equations showed that for large systems the wave propagation is actually suppressed with gain, leading to enhanced localization[16,17] of waves as if the system is absorbing. Subsequently a more rigorous proof[5] for such a symmetry between absorption and amplification was presented for the time-independent wave equation.

Intuitively one would expect that the presence of amplification should facilitate wave propagation, not suppress it, even in disordered systems. At least in the weak scattering (diffusive) regime, amplification always enhances wave transmission.[2,14,15,18] The time-dependent diffusion equation predicts an increased output and a gain threshold above which the system become unstable.[14,15,18] The diffusive description of photon transport in gain media neglects the phase coherence of the wave. Thus it was generally believed that the paradoxical phenomenon may indicate enhanced localization due to interference of coherently amplified multiply reflected waves.[3–6,8,10,11,13] However, amplification of backscattering does not necessarily imply a reduction in transmission because no conservation of the total photon flux is required in gain media. With sufficient gain, wave should be able to overcome losses from backscattering and propagate through the system with increased intensity. To fully understand the origin of this apparent nonintuitive suppression of transmission, we will examine the validity of the solutions derived from the time-independent wave equation which has been commonly employed in describing the wave propagation in active media.

Linearized time-independent wave equations with a complex dielectric constant have been successfully utilized to find lasing modes by locating the poles[19] in the complex frequency plane and to investigate the spontaneous emission noise below the lasing threshold in distributed feedback semiconductor lasers.[20,21] Such equations are known to be inadequate[19,20] to describe the actual lasing phenomena due to their simplicity in dealing with the interactions between radiation and matter. However, it is generally believed that the time-independent equation should suffice to describe the wave propagation in amplifying media, before any oscillations occur. We will unambiguously show that the so-called symmetry[4,5] between amplification and absorption is an artifact due to the unphysical assumption of a finite output in solving the time-independent wave equations, when such solutions cease to exist at large lengths or large gain. We will show that for each system, there is a frequency-dependent

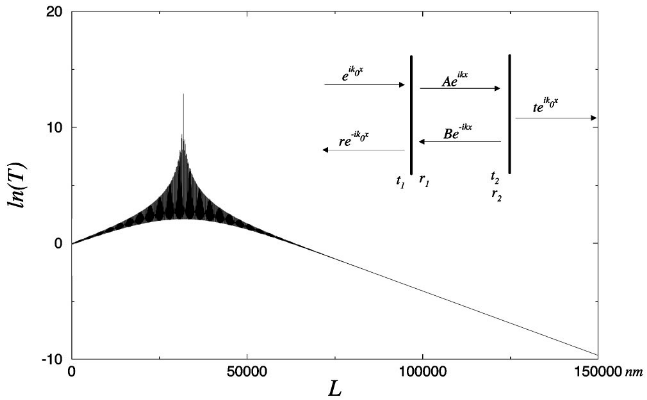

FIG. 1. The logarithm of the transmission coefficient, ln(*T*), versus the length L of a simple Fabry-Pérot setup. The dielectric constant of the Fabry-Pérot device is $\varepsilon=\varepsilon'-i\varepsilon''=2-i0.01$. The dielectric constant of the outside medium is $\varepsilon=1$ without gain. In the inset a schematic representation for the wave transmission in a simple Fabry-Pérot setup is shown.

gain threshold above which no stable time-independent output exists. Solving the time-independent wave equations by incorrectly assuming a fixed output leads to unphysical solutions that do not correspond to the true behavior of the system. These conclusions are supported by numerical solutions of the time-dependent wave equation with a semi-infinite plane wave incident upon a linearly amplifying media.

To see how the unphysical solution could arise from the time-independent equation, we take the simplest example of wave propagation through a uniform active media–the classical Fabry-Pérot setup with the two interfaces acting as feedback mirrors (see the inset in Fig. 1). The time-independent wave solution must satisfy the wave equation,

$$\frac{d^2E(z)}{dz^2}+\frac{\omega^2}{c^2}\varepsilon(z)E(z)=0, \tag{1}$$

where $E(z)$ is the electric field and the dielectric constant $\varepsilon(z)=\varepsilon'(z)-i\varepsilon''(z)$ is complex with the imaginary part signifying amplification from stimulated emission of radiation ($\varepsilon''>0$) or absorption ($\varepsilon''<0$). Here we have assumed a constant ε'' independent of field intensity to represent a uniform linearly amplifying media. We also comment that in electromagnetic theory, the imaginary part of the dielectric constant is proportional to the conductivity of the material and thus cannot be negative. The negative dielectric constant is strictly speaking only a phenomenological way to introduce coherent amplification.[22] Complex potentials known as optical potential have long been employed in nuclear physics to describe inelastic nuclei scattering processes.[23]

For time-independent solutions, the field inside and outside the media are plane waves, with amplitude given as in the inset of Fig. 1. The transmission and the reflection amplitude can be easily obtained by matching the boundary conditions at the two interfaces, with the final expression,

$$t=\frac{t_1t_2e^{ikL}}{1-r_1r_2e^{2ikL}}, \tag{2}$$

where $t_1=2k/(k+k_0)$, $t_2=2k_0/(k+k_0)$, and $r_1=r_2=(k-k_0)/(k+k_0)$ are the effective transmission and reflection coefficients at the left and right two interfaces(mirrors), respectively. $k_0=\sqrt{\varepsilon_0}2\pi/\lambda$ and $k=k'-ik''=\sqrt{\varepsilon'-i\varepsilon''}2\pi/\lambda$ are the wave vectors outside and inside the system, respectively. L is the distance between the ''mirrors.''

The oscillation condition for lasing is correctly given by $1-r_1r_2e^{2ikL}=0$ at which both the transmission and reflection coefficient diverge. However, Eq. (2) also predicts the exponential decrease of the transmission amplitude for large system sizes, asymptotically as $|t|\sim|\exp(-ikL)|=\exp(-k''L)$. This is clearly shown in Fig. 1, where we plot ln(*T*) versus L. Notice that for large L, ln(*T*) decreases as L increases despite the fact that the system has gain. Gain effectively becomes loss at large lengths. Remember the system is homogeneous, thus disorder is definitely not responsible for this strange behavior. We see that the inhibition of wave transmission for large systems is clearly not unique to disordered systems[7] and could not possibly come from the amplification of backscattering.

A more illuminating picture of what is going on can be obtained from the path integral method.[19] In such an approach, the total transmission coefficient can be obtained by adding coherent contributions from all paths of successively reflected and transmitted rays. We obtain a sum of geometrical series,

$$t=t_1t_2e^{ikL}[1+(r_1r_2e^{2ikL})+(r_1r_2e^{2ikL})^2+\cdots]. \tag{3}$$

The first term represents the direct transmission of the incoming wave, and the second term represents the wave which was reflected first by r_2 at the right interface and then by r_1 at the left interface and subsequently transmitted through (see inset of Fig. 1). More terms from sequences of multiple transmissions and reflections at the left and right ''mirrors'' follow. Eq. (3) can be interpreted physically as the output at long times when a continuous plane wave are incident upon the system.

It is a simple matter to verify that Eq. (3) indeed reproduces Eq. (2), provided that the following condition is met:

$$|r_1r_2e^{2ik'L}e^{2k''L}|<1. \tag{4}$$

When the condition given by Eq. (4) is violated, such as when the system size or the gain is large, the output represented by the sum fail to converge, indicating physically the absence of a stable time-independent solution.

The divergence of the transmission above threshold even away from the oscillation pole is the key in understanding the failure of Eq. (2), which is conventionally derived from boundary-condition matching by implicitly assuming that the output is always finite. Normally the physical boundary condition is satisfied when $t_1t_2e^{ikL}+r_1r_2e^{2ikL}t=t$, resulting in Eq. (2). Obviously this condition ceases to be meaningful when t is not finite or ill defined. Consequently, Eq. (2) ceases to represent the physical output. In contrast the condition given by Eq. (4) is always satisfied for lossy media. The nonconvergence of output is a unique phenomenon occurring only to systems with gain. The breakdown of the time-independent wave equation at large gain or large system sizes signals the large fluctuations of the transmission in time and calls for more sophisticated theories that can take into account in more detail the interaction between radiation and matter to correctly describe the response of the system.

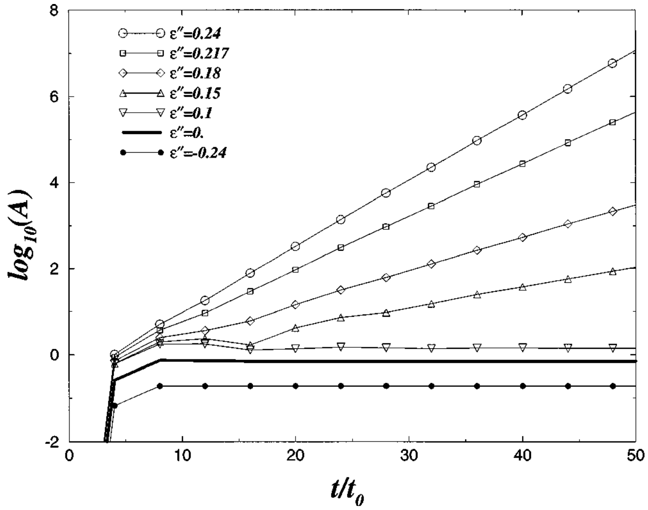

FIG. 2. The logarithm of output at the right side of system versus time of a Fabry-Pérot setup with a plane-wave ($\lambda = 800$ nm) incidence. The length of system is the $L = 4300$ nm and the dielectric constants of the inside and outside materials are $\varepsilon_1 = 9 - i\varepsilon''$ and $\varepsilon_1 = 1$, respectively. The time unit t_0 is the round trip time for the wave to travel through the medium. The critical gain of the system is $\varepsilon''_c = 0.12$.

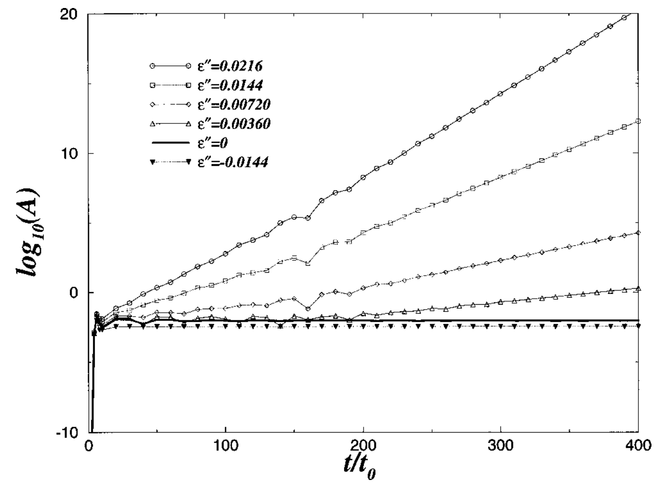

FIG. 3. The logarithm of output at the right side of system versus time of a random system with a plane-wave ($\lambda = 1200$ nm) incidence. The system consists of 50 cells of binary dielectric media with $\varepsilon_1 = 1$ and $\varepsilon_2 = 4 - i\varepsilon''$, respectively. The cell length is fixed at 215 nm and the length of the first medium is given by $a_n = a_0(1 + W\gamma)$, where $a_0 = 95$ nm, $W = 0.8$, and γ is a random number between -0.5 and 0.5. The time unit t_0 is the round trip time for the wave to travel through the system.

To support our picture, we have studied the transmission problem of the simple Fabry-Pérot system directly by solving numerically the time-dependent equation with a well-developed finite-difference-time-domain (FDTD) technique.[24] A semi-infinite planewave of a fixed frequency is incident upon the system from the left. Absorbing boundary conditions have been applied on both ends to eliminate the reflection at the boundaries. We choose $\lambda = 800$ nm and L=4300 nm. The dielectric constant outside is taken to be $\varepsilon_0 = 1$ and the real part of the dielectric constant of the active media is $\varepsilon = 9$. Both mirrors have been chosen to be reflectionless for simplicity (the interfaces provide feedback). For this system, the threshold gain is calculated to be $\varepsilon'' = 0.12$ from Eq. (4). In Fig. 2, we show the field amplitude immediately outside of the active medium as a function of time, for different values of amplification (and absorption). The time unit t_0 here is the round trip time of the light within the media. We expect after some transient time, the transmitted wave should converge to the solution [Eq. (2)] predicted by the time-independent equation. This is indeed what happens when the gain is below the threshold. However, when the gain is above the threshold, the output field appears to increase with time exponentially without bound, indicating the nonexistence of a stable time-independent solution under these conditions.

We have also performed similar calculations on a random system consisting of 50 cells of binary layers with $\varepsilon_0 = \varepsilon_1 = 1$ and $\varepsilon_2 = 4 - i\varepsilon''$. Within the unit cell the thickness of the first layer is a random variable $a_n = a_0(1 + W\gamma)$ where $a_0 = 95$ nm. $W = 0.8$ and γ is a random value in the range $[-0.5, 0.5]$. The cell length is fixed to be 215 nm. The wavelength of the incident wave is $\lambda = 1200$ nm. The results are shown in Fig. 3. Again we see the output field strength increases with time without any bound once the gain reaches some value. This threshold depends not only on frequency but also on disorder configuration of the sample. Figure 3 clearly shows the output always increases with gain for the same input and there is no sign of the suppression of transmission, even in random systems.

These numerical solutions demonstrate unambiguously that for a linearly amplifying medium, there is a frequency-dependent and sample-specific gain threshold above which no physical time-independent solution exists. Solutions from the time-independent equation are valid only below the threshold. This threshold decreases with increasing system size. Unfortunately it is difficult to determine this threshold without explicitly solving the time-dependent equation. One certainly cannot tell from the expression of the total transmission coefficient itself since it is well behaved except exactly at the discrete oscillation poles. One method is to divide the whole system into smaller subsystems and then apply the geometrical sum of multiple reflections and transmission technique, as we have done for the Fabry-Pérot setup, to each subsystem to find the threshold curve for the subsystem. Then the locus formed by all the possible subsystems will be the threshold curve for the whole system. However, this strategy is practical only to simple systems. It is very difficult to carry out such analysis for large or disordered systems. However, since the oscillation poles for the lasing threshold are still correctly given by the divergence of the transmission coefficients from the time-independent solutions, an estimate of the magnitude of the threshold for frequencies away from the pole can be obtained from the threshold values of nearby lasing poles. A study of the distribution of these poles has been carried out for a random layer system by carefully locating the pole position by continuously tuning the gain up at a fixed frequency until the transmission coefficient from the time-independent equation diverges.[9]

The implication of our analysis is that the calculation of transmission and reflection coefficient with the traditional method from time-independent equations become suspect once the gain or system size reaches a certain value. Care has to be taken to ensure that the system is not above the threshold and that the solution is physical. This remark not only applies to studies with transfer matrix[4–8,10,12,13] but also pertains to the application of the powerful *invariant embedding* method[25] which has been widely employed in the studies[3,6,8,11] of transmission problem in random gain media. The presence of a stable time-independent solution is implicitly assumed in these approaches. Consequently, the conclusions of previous studies[3,4,6,8,10,11,13] on the statistical properties of the transmission and reflection coefficient need to be reevaluated in light of the current finding.

In summary, we show that amplification does not suppress wave transmission, as has been claimed previously for random active media. On the contrary, it always enhanced wave propagation. Numerical solution to the time-dependent equation show when the system size (or gain) reaches a frequency- and configuration-dependent threshold, both the amplitude of the reflected and the transmitted wave from a continuous input increases with time without any bound. The erroneous conclusions reached in precious studies was an artifact of the failure of the output wave to converge to a finite value, which must be assumed in any solutions of the time-independent equation. In light of the current findings, some of the conclusions on the statistical properties of the reflection and the transmission coefficient in media with gain become suspect, especially at large system sizes. The time-independent equation is inadequate to describe the amplification of light under these conditions. Nevertheless, the simplicity of the time-independent equation can still be used effectively to locate resonant conditions, even in disordered systems. A complete treatment of the wave propagation in gain media may require the construction of the time-dependent solution out of the continuous and discrete solutions of the time-independent equation.[26] Unlike for the Hermitian system, the completeness of these solutions could not be proved easily when the potential is complex.

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