Random Laser in One Dimension

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We present an analytical approach to random lasing in a one-dimensional medium, consistent with transfer matrix numerical simulations. It is demonstrated that the lasing threshold is defined by transmission through the passive medium and thus depends exponentially on the size of the system. Lasing in the most efficient regime of strong three-dimensional localization of light is discussed. We argue that the lasing threshold should have anomalously strong fluctuations from probe to probe, in agreement with recent measurements.

Random lasing, predicted theoretically by Letokhov [1] more than thirty years ago and discovered experimentally in the past decade [2–5], attracts much attention because of its intrinsic interest and high potential for many practical applications [6]. Theoretical understanding of this phenomenon remains imperfect.

The original approach [1] studied the transport of the disorder-averaged light intensity that can be described by a diffusion formalism. For a random amplifying medium of size L with a mean-free path length of light l_t and gain length l_g the diffusion model predicts that the lasing instability emerges, when the length of the diffusion path L^2/l_t reaches the gain length l_g. Analysis of the interference effects in terms of coherent backscattering corrections [7] does not really modify this criterion. Practically, lasing commences at a different gain than the diffusion model predicts, as seen in both experimental and numerical studies [4,8–10].

The discrepancy between theory and experiment suggests that a physical mechanism, different from diffusive motion, is responsible for lasing from a random medium. Light transport through random media can be described using a discrete sequence of channels between the input and output (see, e.g., [11,12]). Far from the light localization situation the transmissive and reflective properties of a disordered medium can be described by conductive channels. Interference of those channels leads to the average diffusive behavior of the intensity. This picture can be different in amplifying media. In fact each channel α can be crudely characterized by its path length L_α or the travel time τ_α. If we characterize the amplifying properties of the medium by the gain length l_g or by the gain rate $g = v/l_g$ (v is the speed of light inside the medium) then each channel α will show exponential amplification $A_\alpha \propto \exp(L_\alpha/l_g) = \exp(g\tau_\alpha)$. This exponential enhancement selects only channels that have the longest travel time and are not important for the transport in the non-active medium. One possible example of such channels is the ray optics paths forming closed loops [13]. The light trapped in that loop spends much more time inside the medium than the standard diffusion time and therefore experiences larger amplification. These rare channels can be most important in the regime of strong amplification close to lasing. The analysis of these channels differs from the diffusion situation. Solution of the problem for three dimensions is very complicated. In our previous work [9,10] we presented a phenomenological and numerical approach to lasing in ZnO disks showing that even in the extremely open system of resonant scattering particles the fluctuations in scattering channels affect lasing threshold significantly. However, it is important to have an analytical approach that can be used for qualitative understanding of the phenomenon.

In this Letter we study lasing in a one-dimensional (1D) medium where the analytical approach is simplest to develop. A random 1D system will be considered in a model similar to Ref. [8]. This model enables us to identify the relevant channels responsible for lasing as the quasistates (decaying eigenmodes inside the medium) and to compute the lasing threshold in the regime of Anderson localization. We will show that the lasing threshold decreases exponentially with the size of the sample. In fact the photons created inside require an exponentially long time to leave the medium because the transmission is exponentially small in the localization regime. Therefore the exponentially small gain is needed to start lasing.

The results are applicable to higher dimension in the strong localization regime. Actually the regime of strong localization of light is expected to be most efficient for lasing because of the strength of intensity trapping by localized states. There exist at least two experimental studies where strong localization of light has been found [14,15]. Our results can be applied to random lasing in these systems. In particular we predict anomalously strong fluctuations of the lasing threshold from probe to probe in the localization regime. Similar behavior has been discovered in the recent measurements of lasing from the defect states in the forbidden band in liquid crystals [16].

Consider lasing from a 1D scattering medium situated along the x coordinate from $x = 0$ to $x = L$. The medium

is taken to be uniform in two other directions and we restrict our consideration to propagation along the x direction only. Assume that it has a set of scattering particles (or layers), separated from each other by air and located randomly. Practically this characterizes the structure of random layers homogeneous in directions parallel to the interface that can be constructed using random stacking faults in inverted opals or multiple quantum well structures [17].

One can choose an arbitrary point in the middle of the structure $x_s \sim L/2$ and place a source there. Assume the source is located in the air. This setup is relevant for the lasing problem since the source of light of frequency ω is located inside the medium and the generalization for different locations of the source is straightforward. To describe light intensity, one can characterize the left and right halves of the medium by their reflection and transmission coefficients r_l, t_l and r_r, t_r for left and right halves, respectively.

To study random lasing, one needs to introduce the amplification. The easiest way is to assume that the frequency has an imaginary correction $\omega \rightarrow \omega + ig/2$ describing the gain everywhere inside the sample. Then the reflection and transmission coefficients become a function of a complex frequency $r_l(\omega + ig/2)$, $r_r(\omega + ig/2)$. The factor $1/2$ is introduced since the correction to the frequency is responsible for the amplitude gain, while g describes the amplification of the intensity. For the sake of simplicity the gain is taken to be uniform.

The lasing instability takes place when the gain g is sufficiently large so that at some frequency ω the intensity of the light becomes infinity. As we will see, this criterion is equivalent to the singularity in the transmission through the medium suggested in several works as the lasing criterion [8,18], but our approach is more suitable for the real lasers where the source of emission is always located inside the material [4].

The intensity of the emission at the given frequency ω near the source point is defined by the Green function $G(x, x_s)$ of the whole problem. One can reproduce this Green function as the superposition of two plane waves $G(x, x_s) = c_r e^{ik(x-x_s)} + r_r e^{-ik(x-x_s)}$, $x > x_s$; $G(x, x_s) = c_l e^{-ik(x-x_s)} + r_r e^{ik(x-x_s)}$, $x < x_s$; $k = \omega/c$, and it should be continuous at $x = x_s$, while its derivative should have a jump at $x = x_s$ that is set to unity. The coefficients of the Green function can then be expressed as (the requirement to have outgoing waves inside of the medium is applied)

$$c_r = \frac{1}{i\omega}\frac{1 + r_l}{1 - r_l r_r}, \qquad c_l = \frac{1}{i\omega}\frac{1 + r_r}{1 - r_l r_r}. \qquad (1)$$

The lasing instability emerges when the denominator in Eq. (1) reaches zero:

$$r_l(\omega) r_r(\omega) = 1. \qquad (2)$$

Since the total transmission (from the left to the right) through the system can be expressed similar to Eq. (1),

$T = t_r^* t_l/(1 - r_l r_r)$ [11], the criterion Eq. (2) is equivalent to the transmission based criterion [8,18,19].

As we will see the lasing threshold g in the strong localization regime is very small, and an approximate representation of the lasing threshold through the known parameters of the medium without gain is possible. The lasing instability should show up near some maximum of the intensity (or transmission) for the nonactive medium. Consider the very long system without gain having size L much larger than the transport length l_t. In this regime the absolute values of the reflection coefficients r_l, r_r are expected to be close to unity. Therefore the maximum intensity occurs at a frequency where the product of reflection coefficients is a real, positive number. In other words, the sum of reflection phases ($r = |r|e^{i\Phi}$) should be a divisor of 2π,

$$\Phi_r(\omega) + \Phi_l(\omega) = 2\pi n, \qquad n = 0, 1, \ldots. \qquad (3)$$

We denote this condition as resonance. It can be shown that for the closed system (there are 100% reflecting mirrors somewhere at $x < 0$ and $x > L$) the resonances approach the eigenmodes of the whole problem.

Lasing emerges, when the denominator in Eq. (1) becomes zero (divergency of intensity and transmission). Since in the passive medium the absolute values of the reflection coefficient $|r_l|$, $|r_r|$ are less than unity, one needs to add some gain to reach the singularity. At sufficiently small gain, one can use an expansion of phases $\Phi(\omega + ig/2)$ over the gain g in Eq. (2) at the resonance in Eq. (3), where the product of nonamplified reflections is a real number close to unity (similar to the analysis of the absorption effect on the photon lifetimes inside the medium [19]). The expansion yields the lasing condition

$$|r_r r_l| \exp[(g/2)(d\Phi_r/d\omega + d\Phi_l/d\omega)] = 1, \qquad (4)$$

The frequency dependence of the absolute values of the reflection coefficient is neglected here because the large sample length leads to the absolute value of the transmission close to 1 ($1 - |r| \ll 1$). Equation (4) generalizes the standard equation of a laser (cf., e.g., [2,3]) for a 1D disordered medium. The factor in the exponent represents the product of the gain rate and the time that the photon spends trapped inside the medium. Since both reflection coefficients are close to 1 the solution for the lasing threshold can be conveniently expressed in terms of the transmission coefficients t_l, t_r as

$$g_c \approx \frac{|t_r|^2 + |t_l|^2}{d\Phi_r/d\omega + d\Phi_l/d\omega}. \qquad (5)$$

To verify the applicability of the linear expansion we have computed the lasing thresholds for a long 1D chain of 40 scattering particles with refractive index $n = 2$ (size 1 and average interparticle distances equal 1) at frequences between 0.5 and 1.5 (we set the speed of light in vacuum $c = 1$). The methods developed in Ref. [10] have been used and the results have been compared with the prediction [Eq. (5)]. For all our probes we found that Eq. (5) is satisfied with the accuracy of 0.1%. This is not surprising

because the localized modes are not strongly affected by the gain [20].

Equation (5) has a straightforward kinetics interpretation similar to [21]. One can consider the time evolution of the light intensity trapped inside the medium in the localized state (all states are actually quasilocalized because the physical system is open). The inverse sum of phase derivatives represents the attempt frequency to escape out of the trap, while the sum of transmissions is the probability for each attempt to be successful. Then the time evolution for the survival probability p of the intensity inside the media can be written as

$$\frac{dp}{dt} = gp - g_c p. \qquad (6)$$

At $g = g_c$ the gain matches the loss, and the lasing instability emerges. Our consideration fails above the lasing instability, but the predictions for the lasing threshold should be valid. Almost identical arguments can be applied to a localized system of higher dimensionality. The lifetime of excitations trapped by localized modes is similarly defined by the transmissions from the center of the localized state outside of the medium. Since the transmission in the localization regime behaves similar to 1D systems [11] all results remain valid there. Further consideration of the lasing threshold is based on the above analogy. The analysis of contributions of different localized states uses the simple picture of exponential localization inside a lengthy medium. Note that the general problem of transmission through resonances is more complicated but the simplified consideration below remains qualitatively valid. The analysis of the problem in detail will be published elsewhere.

Consider the lasing threshold in the case of uniform gain rate g applied for frequencies within the range $(\omega_0, \omega_0 + \delta\omega)$. Experimentally $\delta\omega$ is very small [4] compared to ω_0 and the average properties of the medium can be considered as invariant within that frequency range. Eigenoptical modes inside the medium corresponding to the resonances [Eq. (3)] can be described by their density of states $\rho(\omega)$ per unit energy and length.

In the regime of the strong localization the transmission decreases exponentially with the size of the sample

$$t \sim \exp(-L/l_1), \qquad (7)$$

where l_1 is the localization length that is close to the transport length l_t. Large transmission fluctuations can be described by the logarithmically normal distribution (see, e.g., [11]) with the standard deviation of the transmission logarithm $\sqrt{L/l_2}$, $l_2 \sim l_t$ (see, however, [22]).

We consider a long system, so that the localization radius l_1 at the reference frequency ω_0 is smaller than the system length, and the number of states (resonances) N_l within the amplified frequency range $\delta\omega$ is large

$$l_1 \ll L, \qquad N_l \sim \rho(\omega_0)L\delta\omega \gg 1. \qquad (8)$$

Each resonance state can then be described by the coordinate of its center x and two transmission coefficients,

with the most significant coordinate dependence given by $t_l(x) \sim \exp(-x/l_1)$ and $t_r(x) \sim \exp[-(L - x)/l_1]$. The gain needed to cause this state to lase Eq. (5) also depends on the reflection phase derivatives. These derivatives describe the dwell times of light sent from the center of the localized state and reflected by the right and left parts of the sample. The distribution of dwell times has been derived for a 1D system in [23]. The most probable time can be estimated as the ratio of the transport length and the characteristic speed of light within the medium $\tau_0 \approx d\Phi/d\omega \sim d(kl_1)/d\omega \sim l_1/v$, while all moments of the distribution diverge for the infinite sample because at large time τ it decreases as $1/\tau^2$ only. Then the lasing threshold for the localized state centered at coordinate x can be estimated as

$$g(x) \sim \frac{1}{\tau_0} \{\exp(-2x/l_1) + \exp[-2(L - x)/l_1]\}. \qquad (9)$$

Lasing occurs when the gain reaches the minimum value of $g(x)$ over all involved states x. This minimum is realized, when the localized state is centered at the middle of the sample $x \approx L/2$:

$$g \propto \exp(-L/l_1). \qquad (10)$$

Thus the lasing threshold depends exponentially on the total length of the system if Eq. (8) is satisfied.

The predicted exponential dependence [Eq. (10)] of the lasing threshold on the system size disagrees with the numerical simulation results [8] for a 1D random system that gives a power law dependence on the localization length (and, accordingly, on the system size L). This discrepancy is probably due to the relatively short lengths L compared to the localization length used in Ref. [8] and the large lasing threshold fluctuations.

The lasing threshold strongly fluctuates from mode to mode due to the variation in the center of localization x [Eq. (9)], transmission [11], and dwell time [23] fluctuations. It can be shown that the lasing threshold distribution is defined by the parameter $N_m = \rho(\omega)\delta\omega\sqrt{Ll_t}$ that is the number of resonant modes in the middle of the sample, having similar transmissions. This distribution changes from the power law distribution at $N_m \ll 1$ to the approximately logarithmically normal distribution at $N_m \gtrsim 1$. In both cases (especially in the first one), the fluctuations of lasing threshold are much larger than its typical value, in agreement with [8]. Strong fluctuations of lasing threshold depending on the pump position, transmission, and dwell times have been recently demonstrated by Vanneste and Sebbah [20] in a numerical analysis of the two-dimensional system. A qualitatively similar interpretation has been suggested there.

Strong fluctuations of lasing threshold are also confirmed by the numerical simulations. The effect of dwell time (phase derivatives) fluctuations [23] of the lasing threshold [Eq. (5)] seems to be less significant than the fluctuations of transmissions possibly due to the special choice of the middle point (the center of resonant states). It

is clear that the transmissions at resonant frequences (and corresponding dwell times) have their own statistics different from that in the off-resonant regime and they are generally much larger. One important consequence is the increase of the sample transparency in the lasing regime since the majority of pumping energy will be distributed between resonant states. This effect has been seen in some thin samples, but there are no systematic studies yet.

Our arguments are directly applicable to several materials including layered or quasi-1D media [14,17] and materials with a 3D localization of light [14,15]. Photonic crystals, where the localized states are formed by the defects, creating excitations within the forbidden band [16], are also of special interest. The pumping frequency for these materials can be chosen far above the localization threshold (or near the pass band [22] for planar layer structures). An interesting alternative opportunity can be reached by placing the dye far from the sample boundary [a preliminary attempt for a TiO_2 based random laser (e.g., [9]) leads to the reduction of the lasing threshold by the factor of 2 for samples very far from Anderson localization]. The characteristic penetration length l_p of the pumping energy can then be made much longer than the localization length at the emission frequency l_1. The lasing threshold will then decrease exponentially with the penetration length, as $\exp(-l_p/l_1)$, until obtaining its minimum value needed to reach finite gain. As discussed above, strong fluctuations of lasing threshold are expected in the regime of Anderson localization. Such fluctuations have been recently observed in a liquid crystal based random lasing system [16].

We have outlined a theory for random lasing in the regime of the strong localization. The lasing threshold decreases exponentially with the size of the system. This can lead to very efficient lasing based on existing [14,15] and possibly new materials with strong localization of light. Random lasing can help one to study the strong localization transition based on the exponential dependence of the lasing threshold on the sample thickness and the huge fluctuations of the lasing threshold that far exceed its typical value.

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