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Light transport and correlation length in a random laser

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A random laser is a strongly disordered, laser-active optical medium. The coherent laser feedback, which has been demonstrated experimentally to be present in these systems beyond doubt, requires the existence of spatially localized photonic quasimodes. However, the origin of these quasimodes has remained controversial. We develop an analytical theory for diffusive random lasers by coupling the transport theory of the disordered medium to the semiclassical laser rate equations, accounting for (coherent) stimulated and (incoherent) spontaneous emission. From the causality of wave propagation in an amplifying, diffusive medium we derive a novel length scale which we identify with the average mode radius of the lasing quasi-modes. We show that truly localized modes do not exist in the system without photon number conservation. However, we find that causality in the amplifying medium implies the existence of a novel, finite intensity correlation length which we identify with the average mode volume of the lasing quasimodes. We show further that the surface of the laser-active medium is crucial in order to stabilize a stationary lasing state. We solve the laser transport theory with appropriate surface boundary conditions to obtain the spatial distributions of the light intensity and of the occupation inversion. The dependence of the intensity correlation length on the pump rate agrees with experimental findings.

1 Introduction

A random laser is a system formed by randomly distributed scatterers embedded in a host medium where either scatterer or host medium or both provide optical gain through stimulated emission first proposed in [1]. Observations of random lasing in a wide variety of systems, like powders of semiconductor nanoparticles [2–5], organic dyes in strongly scattering media [6–8], organic films or nanofibers [9–11] and ceramics [12], have recently triggered a rapidly growing interest in this field. Random lasers share properties with conventional lasers, like threshold behavior [4], narrow spectral lines [16], or photon statistics [17, 18], but also exhibit distinctly different properties like multidirectional emission. Coherent feedback in such systems has unambiguously been demonstrated to be present [17]. It requires the light to be sufficiently confined in the random system. While spatially confined regions from which the laser emission takes place have been observed experimentally [13, 16], the physical origin of coherent feedback, of localized quasimodes and the dependence of their size on the pump rate have remained controversial. The possible theoretical explanations range from preformed random microresonators [19] to multiple random scattering (diffusion) [20], possibly enhanced by self-interference [16] of waves and the resulting onset of Anderson localization (AL) [21]. Conversely, it is an interesting fundamental question how AL of light, which has been understood [22, 23] as a consequence of self-interference, is influenced by the coherent, stimulated amplification in the lasing state. The problem of the intensity distribution in a diffusive random laser

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has been attacked theoretically by phenomenological diffusion models [24] and numerical calculations in one [25,26] and two spatial dimensions [20,27–29]. However, the experimentally observed decrease of the lasing spot size with increasing pump rate, has not been explained so far.

Here we address the issue of the spatially confined lasing spots in diffusive random lasers, and how their size depends on the pump intensity.

2 Model

The propagation of light is described by Maxwell's equations, which may be combined to yield the so-called wave equation for the electric field. Neglecting the polarization degree of freedom we consider in the following the so-called scalar wave equation for the field Ψ, which reads

$$\frac{\omega^2}{c^2}\,\epsilon(\vec{r})\Psi_\omega(\vec{r}) + \nabla^2\Psi_\omega(\vec{r}) = -i\omega\frac{4\pi}{c^2}j_\omega(\vec{r})\ , \tag{1}$$

where c denotes the vacuum speed of light and $j_\omega(\vec{r})$ an external current source. The dielectric constant is $\epsilon(\vec{r}) = \epsilon_b + \Delta\epsilon\, V(\vec{r})$, where the dielectric contrast between the background, ϵ_b, and the scatterers, ϵ_s, has been defined as $\Delta\epsilon = \epsilon_s - \epsilon_b$. The spatial arrangement of the scatterers is described through the function $V(\vec{r})=\sum_{\vec{R}} S_{\vec{R}}(\vec{r}-\vec{R})$, with $S_{\vec{R}}(\vec{r})$ a localized shape function at random locations $\vec{R}$. Linear gain (absorption) is described by a temporally constant, negative (positive) imaginary part of ϵ_b and/or ϵ_s.

In [30–32] we have developed a theory for light transport in disordered media with linear gain or absorption. The result is the diffusion pole structure of the energy-density correlation function in Fourier space $P_{\mathrm{E}}^{\omega}(Q,\Omega)$. In real space $P_{\mathrm{E}}^{\omega}(\vec{r}-\vec{r}\,',t-t')$ describes how the energy density of the light field with frequency ω propagates diffusively between two points in space and time, $(\vec{r},t)$, $(\vec{r}\,',t')$.

The energy-density correlation function is found to be

$$P_{\mathrm{E}}^{\omega}(Q,\Omega) = \frac{N_P}{\Omega + iQ^2D + i\xi_a^{-2}D} \tag{2}$$

where the non-critical numerator N_P is given explicitly in [31] and is not relevant for this discussion. The denominator, however, exhibits the typical diffusion pole structure in a non-conserving system, i. e. featuring a so-called mass term $i\gamma_a = i\xi_a^{-2}D$, which may have either positive or negative sign. The self-consistent solution of the transport theory including self-interference of waves (Cooperon contributions) (see [31,32]) shows that in the presence of absorption or gain the diffusion coefficient D cannot vanish and is in general complex. Therefore, truly Anderson localized modes do not exist in this case.

For the case of linear gain ($\gamma_a < 0$) the wave equation predicts an unlimited growth of the field amplitude and, hence, of the energy density. This means that a stationary lasing state is not possible in this case and, therefore, the limit $\Omega \to 0$ must strictly not be taken in Eq. (2). However, one still can extract the average spot size of a photonic mode from the energy density correlation function. This is done by requiring that the stationary lasing state has been reached *locally*, i. e. within a finite subvolume of the system: Causality requires that the pole of $P_E^{\omega}(Q,\Omega)$, Eq. (2), as a function of Ω resides in the lower complex Ω half-plane. For $\gamma_a < 0$ this is possible only if all the diffusive modes allowed inside a given lasing spot have a wavenumber $Q > Q_{\min} = \sqrt{\mathrm{Re}(-\gamma_a/D)}$. This, in turn, requires that the spot size R_s is

$$R_s = \frac{2\pi}{Q_{\min}} = \frac{2\pi}{\sqrt{\mathrm{Re}(-\gamma_a/D)}}\ . \tag{3}$$

This length is therefore to be identified with the average lasing spot size as observed in experiments. A numerical evaluation is shown in Fig. 1 and agrees well with experimental data, as shown in the inset.

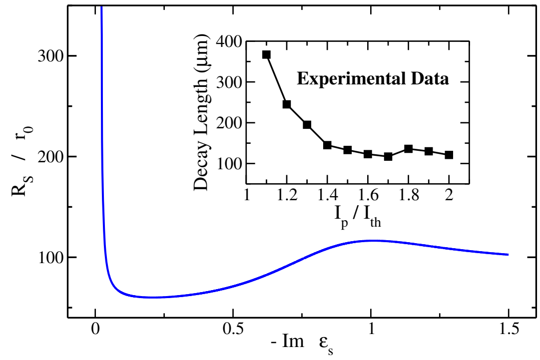

Fig. 1 (online colour at: www.ann-phys.org) The calculated spot size R_s, Eq. (3), in units of the scatterer radius r_0, as explained in the text as a function of the imaginary part of the dielectric constant (pumping) The parameter values used are $\epsilon_b = 1$, $\mathrm{Re}\epsilon_s = 10$, scatterer filling fraction $\nu = 30\%$, light frequency $\omega/\omega_0 = 2.5$. The unit of frequency is $\omega_0 = 2\pi c/r_0$ where c is the vacuum speed of light. The data in the inset are taken from [13] and refer to the spot size of the modes.

3 Lasing and diffusive transport

By considering now a random laser of finite size, causality is intrinsically guaranteed, as will be shown below. In particular, we consider here a three-dimensional random laser model with a homogeneously pumped, active medium which extends infinitely in the (x,y) plane, but has a finite, constant thickness d in the z direction and therefore exhibits a loss of light intensity. The laser-active material is described by the semi-classical laser rate equations, and the light intensity transport by a diffusion equation. In the stationary limit, the laser rate equations can be solved for the population inversion density n_2 to yield

$$n_2 = \frac{\gamma_P}{\gamma_P + \gamma_{21}\left(n_{ph} + 1\right)}. \tag{4}$$

The mean photon number density (light intensity), normalized to N_{tot}, $n_{ph} = N_{ph}/N_{\mathrm{tot}}$, obeys the diffusion equation [24],

$$\partial_t n_{ph} = D_0 \nabla^2 n_{ph} + \gamma_{21}(n_{ph} + 1) n_2, \tag{5}$$

where the last term on the r.h.s. describes the intensity increase due to stimulated and spontaneous emission, as described by the semi-classical laser rate equations. Since in the slab geometry described above ensemble-averaged quantities are translationally invariant in the (x, y) plane, but not along the z direction, a Fourier representation in the (x, y) plane in terms of $n_{ph}(\vec{Q}_{||}, z)$, $n_2(\vec{Q}_{||}, z)$ is convenient,

$$\partial_t n_{ph} = -D_0 Q_{||}^2 n_{ph} + D_0 \partial_z^2 n_{ph} + \gamma_{21} \int \frac{d^2 Q'_{||}}{(2\pi)^2} n_{ph}(\vec{Q}_{||} - \vec{Q}'_{||}, z) n_2(\vec{Q}'_{||}, z) + \gamma_{21} n_2 \tag{6}$$

In the hydrodynamic limit for stationary lasing, the photon density response function $P(\vec{Q}_{||}, z, \Omega)$ may be constructed from that to yield

$$P_E(\vec{Q}_{||}, z, \Omega) = \frac{i\gamma_{21}}{\Omega + iQ_{||}^2 D_0 + i\xi^{-2} D_0} \qquad \text{where} \qquad \xi = \sqrt{\frac{D_0}{\gamma_{21}} \frac{n_{ph}}{n_2}} \tag{7}$$

where the correlation length ξ is now a real and positive quantity, hence ensuring the causality of the diffusion pole for this finite size system.

Numerical evaluations of the photon density as a solution of the diffusion equation with appropriate boundary conditions as well as a numerical evaluation of the correlation length ξ are shown in Fig. 2. In the left panel of Fig. 2 the mean photon number displays a monotonically increasing behavior with increasing pumping at sample surface, changing into a linear behavior as observed experimentally [13]. Above threshold the correlation length (spot size) exhibits a decreasing behavior, which we numerically analyzed to follow an inverse square-root law.

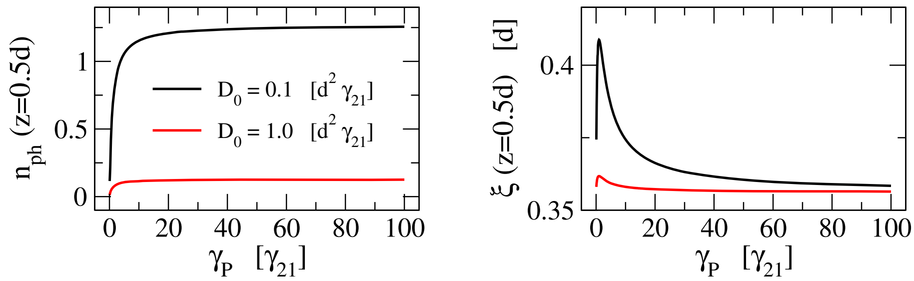

Fig. 2 (online colour at: www.ann-phys.org) The left panel displays the mean photon number as a function of the pumping rate P at the film surface, whereas the right panel features the correlation length (lasing spot size).

4 Conclusion

We discussed the possibility to use linear response theory of disordered media with linear gain, including self-interference (Cooperon) contributions, to extract the mean spot size of lasing modes in random lasers. Good agreement with experiments is found. Furthermore, we presented a theory of random lasing for systems of finite size. In the considered slab geometry, we also calculated the correlation length, and found here that above threshold the spot size decreases with increasing pumping P, as found in experiments. We analyzed this decay to be of the form $\propto 1/\sqrt{P}$, which is open to experimental tests.

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