IOP PUBLISHING JOURNAL OF OPTICS A: PURE AND APPLIED OPTICS

J. Opt. A: Pure Appl. Opt. **11** (2009) 114012 (6pp) doi:10.1088/1464-4258/11/11/114012

# Light transport and localization in diffusive random lasers

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Received 16 February 2009, accepted for publication 13 July 2009
Published 16 September 2009
Online at stacks.iop.org/JOptA/11/114012

**Abstract**

We develop an analytical theory for diffusive random lasers by coupling the transport theory of the disordered medium to the semi-classical 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 further that loss at the surface of the laser-active medium is crucial for stabilizing a stationary lasing state. The solution for the transport theory of random lasers for a layer geometry with appropriate surface boundary conditions yields the spatial profile of the light intensity and of the population inversion. The dependence of the intensity correlation length on the pump rate is in qualitative agreement with experimental and numerical findings.

**Keywords:** wave propagation in random media, random lasers, disordered media, light localization

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

A random laser is a system formed by randomly distributed scatterers embedded in a host medium where the scatterer or the host medium or both provide optical gain through stimulated emission [1]. Recent 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 triggered a rapidly growing interest. For reviews with comprehensive lists of references see [13, 14]. Random lasers share some properties with conventional lasers, like threshold behavior [4], narrow spectral lines [15], and photon statistics [16, 17], but also exhibit distinctly different properties like multidirectional emission. Coherent feedback has unambiguously been demonstrated to be present in strongly disordered random lasers [16]. 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 [15, 13], the physical origin of coherent feedback and of localized quasi-modes, and the dependence of their size on the pump rate, have remained controversial. The possible theoretical explanations range from preformed random microresonators [18] to multiple random scattering (diffusion) [19], possibly enhanced by self-interference [15] of waves and the resulting onset of Anderson localization (AL) [20]. Conversely, it is an interesting fundamental question how AL of light, which has been understood [21, 22] 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 has been attacked theoretically using phenomenological diffusion models [23] and numerical calculations in one [24, 25] and two spatial dimensions [26–28, 19]. However, the experimentally observed decrease of the lasing spot size with increasing pump rate has not been explained so far.

In this paper, we address the question of the size of lasing spots in diffusive random lasers using an analytical transport theory. We begin the analysis in section 2 by giving an outline of the transport theory of light in a disordered medium with linear gain (i.e., constant-in-time amplification rate), including self-interference (so-called cooperon) contributions. By a phenomenological analysis we show that causality implies in the presence of linear gain a novel length scale which is to be identified with the average radius (spot size) of a lasing mode, $R_s$. In section 3 we extend the theory for a system with linear gain to a transport theory including non-linear gain due to stationary lasing above threshold. Observing that a stationary lasing state is possible only if the amplification in the medium is compensated by loss at the surface of the system, we consider a model for a finite-size random laser with infinite extent in the $(x, y)$ plane, but finite, constant thickness in the $z$ direction, a geometry relevant for many experimental systems [3, 4, 15, 16]. Coupling the diffusive transport theory to the rate equations of a four-level laser in the stationary state we derive an analytical expression for the intensity correlation length $\xi$, to be identified with the average lasing spot size, $R_s$. Due to the surface boundary conditions the spatial extent of a lasing spot in the $(x, y)$ plane obtains a $z$-dependent profile, $\xi(L)$. We also analyze its dependence on the pump rate. The conclusions are drawn in section 4, on the pump rate and on the depth of the mode along the $z$ direction.

## 2. Transport theory for a diffusive, linear gain medium and causality

The propagation of light is described by its wave equation. Neglecting the polarization degree of freedom we consider in the following the scalar wave equation for the field $\Psi$. It reads

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

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, $\epsilon_b$, and the scatterers, $\epsilon_s$, 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 $\vec{R}$. The linear gain (absorption) is described by a temporally constant, negative (positive) imaginary part of $\epsilon_b$ and/or $\epsilon_s$.

In [29–31] we have developed a theory for light transport in disordered media with linear gain or absorption. It results in an energy–density correlation function $P_E^\omega(\vec{r} - \vec{r}', t - t')$, which describes how the energy density of the light field with frequency $\omega$ propagates diffusively between two points in space and time, $(\vec{r}, t)$, $(\vec{r}', t')$. The Fourier transform of the energy–density correlation function $P_E^\omega(Q, \Omega)$ is obtained as

$$P_E^\omega(Q, \Omega) = \frac{N_P}{\Omega + \mathrm{i}Q^2 D + \mathrm{i}\xi_a^{-2} D} \qquad (2)$$

where the expression for the coefficient $N_P$ is given explicitly in [30], but is not relevant for the present purpose. The denominator of equation (2) exhibits the expected diffusion pole structure with the diffusion coefficient $D$. In addition, in the case of a non-conserving medium, i.e. net absorption (gain), there appears the (purely imaginary) term $\mathrm{i}\gamma_a = \mathrm{i}\xi_a^{-2}D$, which has a positive (negative) imaginary part and does not vanish in the hydrodynamic limit, $\Omega \to 0$, $Q \to 0$. The self-consistent solution of the transport theory including self-interference of waves (cooperon contributions; see [30, 31]) shows that in the presence of absorption or gain the diffusion coefficient $D$ cannot vanish and is in general complex. Hence, truly Anderson localized modes do not exist in this case.

For the case of absorption ($\gamma_a > 0$) it is seen by Fourier transforming equation (2) w.r.t. time, $P_E^\omega(Q, t) = \mathrm{i}N_P\, \mathrm{e}^{-(Q^2 D + \gamma_a)t}$, that $\mathrm{Re}\,\gamma_a$ represents the loss rate of the photonic energy density due to absorption in the medium. Fourier transforming equation (2), on the other hand, w.r.t. space in the stationary limit ($\Omega \to 0$) shows that $\xi_a = \mathrm{Re}\sqrt{(\gamma/D)}$ is the length scale over which the energy density of diffusive modes is correlated in the lossy medium.

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 equation (2). Such a behavior of linear gain is expected only during the exponential intensity growth shortly after the onset of lasing. A complete theory of random lasing must, therefore, take into account either the full temporal dynamics of the system, or in a stationary state additional surface loss effects must compensate for the gain in the medium (see section 3). Nevertheless, we can extract a characteristic size of a stationary lasing spot from this theory 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 $P_E^\omega(Q, \Omega)$, equation (2), as a function of $\Omega$ resides in the lower complex $\Omega$ 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 is

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

This is the characteristic, maximal size of a spatial region over which diffusive modes can be causally correlated in the stationary lasing state. We conjecture that, hence, this size is to be identified with the lasing spot size observed experimentally [15, 13] in diffusive random lasers. Since according to the microscopic theory [30] the growth rate $(-\gamma_a)$ is, for small linear gain, proportional to the average gain in the medium, $\gamma_a \propto \mathrm{Im}\,\overline{\epsilon(\vec{r})}$, we predict the spot size to be inversely proportional to the gain.

More generally, despite the fact that the linear gain assumption is not suited to describe stationary lasing, it can be used to estimate the laser threshold, i.e. the critical pump rate for lasing. Amazingly, this is a rather general remark. For example, in a simpler system of a single microsphere with gain, it has been shown [32] that the scattering coefficients calculated within linear response lose their causality just at the point where the sphere crosses its lasing threshold. Applied to our random laser system, this means that the threshold for lasing within a spot of size $R_s$ is reached when the transport coefficient $-\gamma$, determined by the pump rate via the microscopic transport theory [30, 31], reaches the value given by equation (3).

In figure 1 we show the numerical evaluation of the spot size $R_s$ as a function of increasing $\mathrm{Im}\,\epsilon_s$ for typical parameters, as given in the figure caption. The imaginary part of the dielectric constant is a measure of external pumping, since

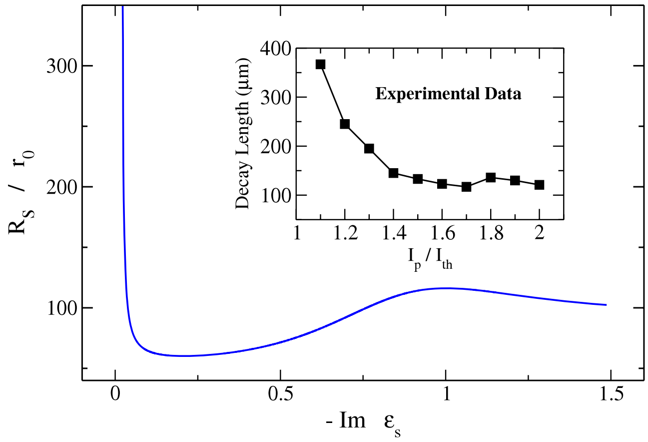


**Figure 1.** The spot size $R_s$, equation (3), in units of the scatterer radius $r_0$, as obtained by causality considerations (see the text) as a function of the imaginary part of the dielectric constant of the scatterers. The parameter values used are $\epsilon_b = 1$, $\mathrm{Re}\,\epsilon_s = 10$, scatterer filling fraction $\nu = 30\%$, light frequency $\omega/\omega_0 = 2.5$. The light 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.

the gain is given by the population inversion of the laser. Therefore, larger pumping yields higher inversion and leads to a larger $\mathrm{Im}\,\epsilon_s$. The calculated spot size displays a qualitative agreement with the experimental data [13] (spot size versus pump intensity/threshold intensity) shown in the inset.

## 3. Transport theory of random lasing

As remarked in section 2, a stationary lasing state in a homogeneously pumped system is possible only if the system is finite, so that surface loss effects can compensate the gain in the medium. To avoid the causality problem, 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. The laser-active material is described by the semi-classical laser rate equations, and the light intensity transport by a diffusion equation. In particular, the rate equations for a four-level laser are

$$\frac{\partial N_3}{\partial t} = \frac{N_0}{\tau_P} - \frac{N_3}{\tau_{32}} \tag{4}$$

$$\frac{\partial N_2}{\partial t} = \frac{N_3}{\tau_{32}} - \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}}\right) N_2 - \frac{(N_2 - N_1)}{\tau_{21}} n_{ph} \tag{5}$$

$$\frac{\partial N_1}{\partial t} = \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}}\right) N_2 + \frac{(N_2 - N_1)}{\tau_{21}} n_{ph} - \frac{N_1}{\tau_{10}} \tag{6}$$

$$\frac{\partial N_0}{\partial t} = \frac{N_1}{\tau_{10}} - \frac{N_0}{\tau_P} \tag{7}$$

$$N_{tot} = N_0 + N_1 + N_2 + N_3, \tag{8}$$

where $N_i = N_i(\vec{r}, t)$, $i = 0, 1, 2, 3$, are the population number densities of the corresponding electron level ($i \in \{1, \ldots, 4\}$), $N_{tot}$ is the total number of electrons participating in the lasing process, $\gamma_{ij} \equiv 1/\tau_{ij}$ are the rates of transition from level $i$ to $j$, and $\gamma_{nr}$ is the non-radiative decay rate of the laser level 2. $\gamma_P \equiv 1/\tau_P$ is the rate of transition due to homogeneous, constant, external pumping. Furthermore $n_{ph} \equiv N_{ph}/N_{tot}$ is the photon number density, normalized to $N_{tot}$. In the stationary limit (i.e. $\partial_t N_i = 0$), the above system of equations can be solved for the population inversion $n_2 = N_2/N_{tot}$ to yield (with $\gamma_{32}$ and $\gamma_{10}$ assumed to be large compared to all other rates)

$$n_2 = \frac{\gamma_P}{\gamma_P + \gamma_{nr} + \gamma_{21}(n_{ph} + 1)}. \tag{9}$$

The photon number density (light intensity), normalized to $N_{tot}$, $n_{ph} = N_{ph}/N_{tot}$, obeys the diffusion equation [23],

$$\partial_t n_{ph} = D_0 \nabla^2 n_{ph} + \gamma_{21}(n_{ph} + 1) n_2, \tag{10}$$

where the last term on the rhs describes the intensity increase due to stimulated and spontaneous emission, as described by the semi-classical laser rate equations. Since in the slab geometry 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,

$$\partial_t n_{ph} = -D_0 Q_{||}^2 n_{ph} + D_0 \partial_z^2 n_{ph} + \gamma_{21} \int \frac{d^2 Q'_{||}}{(2\pi)^2} n_{ph}(\vec{Q}_{||} - \vec{Q}'_{||}, z) n_2(\vec{Q}'_{||}, z) + \gamma_{21} n_2. \tag{11}$$

We now seek the photon density response function $P(\vec{Q}_{||}, z, \Omega)$, which describes the response of the photon density, $n_p h$, to the distribution of the population inversion, $n_2$, in order to determine the transport coefficients. In the stationary case ($\partial_t n_{ph} = 0$) and in the long-wavelength limit along the $(x, y)$ plane ($Q_{||} \to 0$), the $z$ derivative in equation (11) can be expressed without derivatives in terms of $n_{ph}$ and $n_2$ only. Plugging this back into equation (11) yields

$$\left[\partial_t + D_0 Q_{||}^2 + \frac{\gamma_{21} n_2}{n_{ph}}\right] n_{ph}(\vec{Q}_{||,z}, t) = \gamma_{21} n_2(\vec{Q}_{||,z}, t) \tag{12}$$

and, hence, after Fourier transforming w.r.t. time, the diffusion form of the density response function,

$$P_E(\vec{Q}_{||}, z, \Omega) = \frac{i\gamma_{21}}{\Omega + iQ_{||}^2 D_0 + i\xi^{-2} D_0}, \tag{13}$$

where from equation (12) the correlation length $\xi$ is defined as the real, positive quantity

$$\xi = \sqrt{\frac{D_0}{\gamma_{21}} \frac{n_{ph}}{n_2}}. \tag{14}$$

As seen from equation (13) the pole structure of $P_E$ in this finite-size, diffusive system is perfectly causal. The square of the correlation length $\xi$ remains positive, indicating an effective loss out of a given $Q_{||}$ mode. This is due to the loss of intensity at the surfaces. Additionally, the mass term becomes less and less significant as the laser intensity in the sample builds up, because the relative population inversion clearly obeys $n_2 \leqslant 1$ whereas the relative photon number is not restricted.

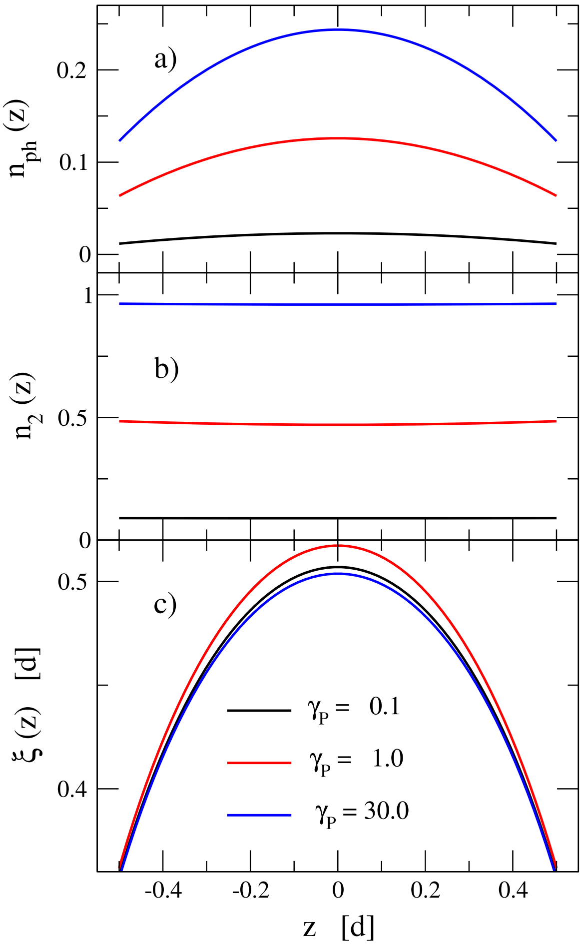


**Figure 2.** The following quantities are shown as a function of $z$ for different values of the pump rate $\gamma_P$: (a) the photon number, which increases monotonically with increasing pump rate and has its maximum in the film center ($z = 0$); (b) the population inversion, which is inversely proportional to $n_{\mathrm{ph}}$—see equation (9); (c) the correlation length (spot size), which clearly behaves non-monotonically with increasing pumping. For all panels the diffusion constant is $D_0 = 1\,d^2\gamma_{21}$.

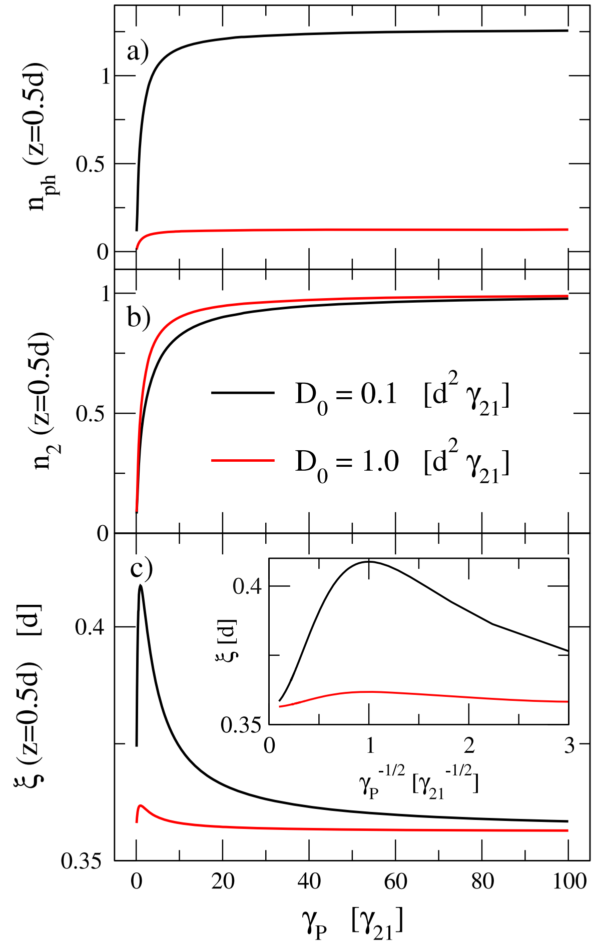


**Figure 3.** The figure shows the following quantities as a function of the pumping rate $P$ at the film surface ($z = \pm 0.5d$): (a) the photon number which is displaying saturation for strong pumping; (b) the population inversion, also saturating; (c) the correlation length (spot size) showing non-monotonic behavior. The inset of panel (c) displays the $1/\sqrt{\gamma_P}$ dependence of the intensity correlation length $\xi$ on the pump rate above threshold ($\gamma_P > 1$; see the discussion in the text).

Since for homogeneous pumping the averaged photon density does not depend on $x$ or $y$, equation (11) simplifies in the stationary limit to

$$D_0 \partial_z^2 n_{\mathrm{ph}} = -\gamma_{21}(n_{\mathrm{ph}} + 1) n_2 \tag{15}$$

and $n_{\mathrm{ph}}(z)$ is finally determined via equation (9) by the regular differential equation

$$\partial_z^2 n_{\mathrm{ph}}(z) = -\frac{\gamma_{21}}{D_0} \frac{(\gamma_P/\gamma_{21})}{1 + \frac{(\gamma_P/\gamma_{21})}{n_{\mathrm{ph}}(z)+1}}. \tag{16}$$

Equations (16), (9) and (14) comprise the complete description of the spatial photon density profile perpendicular to the lasing film and the intensity correlation length (spot size) parallel to the film.

Numerical evaluations of equations (16), (14) and (9) are shown in figures 2 and 3. In figure 2 the photon number $n_{\mathrm{ph}}(z)$, population inversion $n_2(z)$ and correlation length $\xi(z)$ are shown as functions of $z$ for different values of external pumping, characterized by the pumping rate $\gamma_P$. The value of the diffusion constant was chosen to be $D_0 = 1\,d^2\gamma_{21}$, where $d$ is the width of the film. In panel (a) of figure 2 the photon number displays a monotonically increasing behavior with increasing pumping. The maximum of the intensity resides in the center of film ($z = 0$), since this is the position farthest from the boundaries, and therefore with lowest loss of intensity. The population inversion, equation (9), behaves inversely to $n_{\mathrm{ph}}(z)$; see equation (9). In contrast to this rather expected behavior, the correlation length $\xi(z)$ as given by equation (14) exhibits a non-monotonic behavior with increasing pumping. For pumping rates $\gamma_P < \gamma_{21}$ the correlation length increases, but for pumping rates $\gamma_P > \gamma_{21}$, $\xi$ is decreasing. The equality between $\gamma_P$ and $\gamma_{21}$ marks the situation where electrons are excited into the upper laser level as fast as they relax to lower levels. Therefore this characterizes the lasing threshold. Available experimental data [15, 13] also indicate a decreasing behavior of the spot size above threshold. Measurements of the intensity correlation length below threshold have not yet been reported.

The same quantities are shown in figure 3 as a function of external pumping $\gamma_P$ at the surface of the random laser. Photon number and population inversion both display saturation behavior. Panel (c) of figure 3, however, exhibits the non-monotonic behavior of the correlation length. This plot is to be directly compared to experimental data [13], as e.g. shown in the inset of figure 2. There is a good qualitative and even quantitative agreement between calculated and measured spot size. The inset of panel (c) shows that the dependence of the spot size $\xi$ on the pump rate $\gamma_P$ above threshold ($\gamma_P$) is predicted to be

$$\xi(\gamma_P) = \xi(\infty) + \alpha/\sqrt{\gamma_P} \tag{17}$$

with a proportionality constant $\alpha$. This result is open for further experimental tests.

## 4. Conclusion

We have discussed how a linear response theory for light transport, including self-interference effects, in disordered media with linear gain predicts threshold behavior of the intensity. Even more interestingly it also predicts a characteristic, average radius of lasing modes, dictated by causality. We identify this length scale with the spot size of the random laser as measured in experiments [15, 13] and find qualitatively good agreement. Further, we have proposed an analytical transport theory for random lasing in finite systems. A finite system size is necessary for surface loss to compensate the gain in the medium and, hence, to stabilize a stationary lasing state. In particular, we consider a slab geometry where in the medium the light intensity propagates diffusively and the loss through the surfaces is included by appropriate boundary conditions. The theory allows for the first time for an analytical calculation of the intensity correlation length of this system, describing the spatial extent of a mode (spot size). The spot size is predicted to behave non-monotonically as a function of external pumping, i.e. increasing below and decreasing above the laser threshold. A comparison with experiments reveals qualitatively good agreement. Our prediction of its functional dependence on the pump rate is open to further experimental tests.

Our future work will include solving for semi-analytical light transport theory with self-interference contributions when the system is self-consistently coupled to the laser rate equations.

## Acknowledgments

Useful discussions with K Busch, H Cao, D Chigrin, P Henseler, B Shapiro, C M Soukoulis and M Trujillo Martinez are gratefully acknowledged. This work was supported in part by the Deutsche Forschungsgemeinschaft through grant No. KR1726/3 (RF, JK) and FG 557.

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