Random lasing in disordered arrays of ZnO nanorods

Regine Frank^a and Andreas Lubatsch^b

^a Karlsruhe Institute of Technology (KIT), Institut für Theoretische Festkörperphysik, Wolfgang-Gaede-Straße 1, 76131 Karlsruhe, Germany

^b Rheinische Friedrich-Wilhelms-Universität Bonn, Physikalisches Institut and Bethe Center for Theoretical Physics, Nussallee 12, 53115 Bonn, Germany

ABSTRACT

A diffusive theory of random lasing is derived for finite systems comprised of disordered arrays of laser active ZnO nanopillars. The diffusive transport of the light intensity is coupled to the semiclassical laser rate equations, therefore incorporating nonlinear optical gain in this effectively two dimensional system. We solve the resulting boundary condition problem to obtain the full spatial intensity profile of lasing spots in dependence of the pumprate and other system parameters. Our theory predicts two different types of random laser modes in effectively two dimensional systems in general. A surface mode with a large size extending over the entire sample width, and a bulk mode, with small laser spot sizes. We discuss their origin and characteristics.

Keywords: Random lasing, disordered systems, light propagation, active media, light localization

1. INTRODUCTION

A random laser is a most intriguing system composed by randomly positioned scatterers surrounded by a socalled host medium, where optical gain is provided by the scatterer, the host or both. Even in the absence of a resonator, stimulated emission and lasing action occurs as was first pointed out already in 1968.¹ A vast amount of experimental work in a range of physical systems ranging from powders of semiconductor nanoparticles,²⁻⁶ to ceramics,⁷ to organic laser dyes placed in strongly scattering media,⁸⁻¹⁰ to organic films or nanofibers,¹¹⁻¹⁴ results in a spiraling growth of interest. Among the many reviews available on this subject the reader is referred to references^{15,16} and references therein. Random lasers share some properties with conventional lasers, like threshold behavior,³ narrow spectral lines,¹⁷ or photon statistics,^{18,19} but also exhibit distinctly different properties like multidirectional emission. Coherent feedback has unambiguously been demonstrated to be present in strongly disordered random lasers.¹⁸ 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,^{15,17} 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 microresonators²⁰ to multiple random scattering (diffusion),²¹ possibly enhanced by self-interference¹⁷ of waves and the resulting onset of Anderson localization $(AL)^{22}$ of light. Conversely, it is an interesting fundamental question how AL of light, which has been understood^{23,24} as a consequence of self-interference, is influenced by the coherent, stimulated amplification in the lasing state.^{25,26} The problem of the intensity distribution in a diffusive random laser has been attacked theoretically by phenomenological diffusion models^{27,28} and numerical calculations in one^{29,30} and two spatial dimensions.^{21,31–33} 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 occurrence of stationary lasing states in an effectively two dimensional system of disordered nanorods. Our interest is especially focused on the average lateral extent of the observed lasing spots under uniform pumping when the system is operated in the diffusive regime. This paper is organized as follows. First, we discuss the model and the transport theory with its governing equations describing the propagation of a light field in this non-conserving radom medium. Then, we very briefly state main results

Active Photonic Materials IV, edited by Ganapathi S. Subramania, Stavroula Foteinopoulou, Proc. of SPIE Vol. 8095, 80950R · © 2011 SPIE · CCC code: 0277-786X/11/\$18 · doi: 10.1117/12.893976

Further author information: (Send correspondence to R. Frank)

R. Frank: E-mail: rfrank@tfp.physik.uni-karlsruhe.de



Figure 1. Displayed is a schematic setup of the described system, consisting of nanorods composed of laser active ZnO. These nanorods are in our model assumed to be identical but otherwise placed at random locations. The surrounding medium may be vacuum as well as an dielectric medium, including absorption and optical gain. This system is uniformly pumped (not depicted in the sketch) upon which the occurrence of lasing from certain regions in the sample may be observed. This is shown as the bright magenta spot. These lasing spots occur at random position, with a mean size depending on system parameters, such as nanorods size and density per volume, pump strength and intensity leakage to the environment.

known from the semiclassical theory of lasing, which is based on the the consideration of rate equations, as we will need them here later. In section 2 we introduce a diffusion theory for light intensity which includes the non-linear gain due to the stationary lasing above threshold from the laser rate equations. In general, a stationary lasing state may only occur if the intensity amplification within the medium is compensated by an appropriate loss, usually through the surfaces of the finite sized systems. Therefore, we assume a two dimensional model which is finite in y direction and infinite in the x-direction, infinite as compared to both the wave length of the light and light intensity related length scales. This geometric setup is relevant for experimental systems.^{5,34-36} We derive for such systems an analytical expression for the light intensity correlation length by coupling the laser rate equations to the 2d diffusive transport theory. This correlation length describes the area in the sample in which the lasing intensity builds up and hence is an expression for the average lateral size of the lasing spots in the disordered sample. Due to the boundary conditions at the sample's surface the spatial extention of a lasing spot in the x direction obtains a y-dependent profile. We also analyze its dependence on the pump rate. Furthermore, we discuss the occurrence of two different types of lasing modes. The detailed summary and conclusions are finally presented in section 3 and 4.

2. MODEL AND TRANSPORT THEORY

We consider a random laser model with a uniformly pumped, active medium which is schematically shown in Fig. 1. It consists of nanorods composed of laser active ZnO. These nanorods are in our model assumed to be identical cylinders but otherwise placed at random locations, forming the actual random medium. The surrounding hostmedium may be vacuum as well as any dielectric medium, possibly also including absorption and optical gain. This system is uniformly pumped, as stated above (not depicted in the sketch) upon which the occurrence of lasing from certain regions in the sample may be observed. We consider the TM modes (E-polarization) having their electric vector perpendicular to the PC plane, and TE modes (H-polarization) having their magnetic vector perpendicular to the PC plane. In a two dimensional disordered photonic crystal (2d PC) setup the wave equation for the three dimensional electric field $\vec{E}(\vec{r})$ as derived from Maxwell's equations can

be written as

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E}(\vec{r}) - \frac{\omega^2}{c^2} \epsilon(\vec{r}) \vec{E}(\vec{r}) = 0 \tag{1}$$

where a Fourier transform from time t to light frequency ω has been applied. Furthermore, $\epsilon(\vec{r})$ is the dielectric function of the system, describing the random arrangement of the scattering cylinders embedded in the hostmedium. For a TM-polarized wavefield which is here characterized by $\vec{E}(\vec{r}) = E_z(x, y)$, the divergence $\nabla \cdot \vec{E}(\vec{r})$ is always zero. Therefore, by using the identity $\nabla \times \nabla \times \vec{C} = \nabla(\nabla \cdot \vec{C}) - \Delta_3 \vec{C}$ where Δ_3 is the 3d Laplacian, we may write

$$\Delta_2 E_z(x,y) + \frac{\omega^2}{c^2} \epsilon(x,y) E_z(x,y) = 0$$
⁽²⁾

where $\Delta_2 = \partial_x^2 + \partial_y^2$ is the 2d Laplacian and the equation is now scalar. Throughout this paper we will from now on neglect the subscripts and write the 2d scalar wave equation according to

$$\Delta E(x,y) + \frac{\omega^2}{c^2} \epsilon(x,y) E(x,y) = 0.$$
(3)

The dielectric constant is $\epsilon(x, y) = \epsilon_b + \Delta \epsilon V(x, y)$, 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 ϵ_s . From the scalar wave equation for the TM mode electric field Eq. (3), we deduce the 2d disorder averaged retarded Green's function in Fourier space $G_{\vec{q}}^R(\omega)$

$$G_{\vec{k}}^{R}(\omega) = \left(\frac{\omega^{2}}{c^{2}}\epsilon_{b} + q^{2} + \Sigma(\omega)\right)^{-1}$$

$$\tag{4}$$

where $\Sigma(\omega)$ is the selfenergy originating in the scattering off the random potential $\epsilon(x, y)$.

In order to consider the light intensity, one needs, however, to consider the so-called two-particle Green's function, here in position space

$$\langle G^{R}(\vec{r}_{1},\vec{r}_{1}\,';\omega_{1})G^{A}(\vec{r}_{2},\vec{r}_{2}\,';\omega_{2})\rangle =:\Gamma^{\omega_{1},\omega_{2}}(\vec{r}_{1},\vec{r}_{1}\,';\vec{r}_{2},\vec{r}_{2}\,').$$
(5)

As the geometry of the nanorod system is effectively of two dimension in which one dimension, say x is unbounded, and the other, say y is not, we apply a Fourier transform in the x-direction, already when changing from spatial coordinates $(\vec{r}_1, \vec{r}_1', \vec{r}_2, \vec{r}_2')$ to the according center of mass and relative coordinates. For the ydirection, after switching to center of mass and relative coordinates, we apply a Fourier transform to the relative component only. This is justified since, we assume the lateral extent of the sample larger than the wavelength of the light. The corresponding center of mass component, describing the scale over which the light intensity changes, is not Fourier transformed. Then solving the corresponding equation of motion for Γ yields an expression for the two dimensional energy density $P^{\Omega}(Q_x, y)$ depending on the coordinate y, and Ω is the center of mass frequency.²⁵⁻²⁷

In the following we develop a purely diffusive approximation to the above outlined theory. Before we detail our actual method, we have to first revisit the lasing itself.

2.1 Lasing

To describe lasing behavior in general it has been proven helpful to apply the semiclassical laser theory based on rate equations. For relevant materials, we consider a four level laser as depicted in Fig. 2. In this scheme, electrons are (constantly) pumped with a given pump rate γ_P from the ground state E_0 to the highest involved energy state E_3 which has a short relaxation time τ_{32} , therefore, electrons quickly relaxate to the upper laser level E_2 . The transition from E_2 to E_1 is the so-called laser transition, in which laser photons with energy $\hbar\omega$



Figure 2. Displayed is a schematic four level laser system. Electrons are (constantly) pumped with pump rate γ_P from the ground state E_0 to the highest state E_3 which has a short relaxation time τ_{32} so electrons quickly relaxate to the upper laser level E_2 . The transition from E_2 to E_1 is the so-called laser transition, in which photons with energy $\hbar\omega$ are emitted. Finally, electrons leave the lower laser level E_1 quickly towards the ground state.

are emitted. Finally, electrons leave the lower laser level E_1 quickly towards the ground state, where they are pumped again. The appropriate rate equations for this just described laser system are given as

$$\frac{\partial N_3}{\partial t} = \frac{N_0}{\tau_P} - \frac{N_3}{\tau_{32}} \tag{6}$$

$$\frac{\partial N_2}{\partial t} = \frac{N_3}{\tau_{32}} - \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}}\right) N_2 - \frac{(N_2 - N_1)}{\tau_{21}} n_{ph}$$
(7)

$$\frac{\partial N_1}{\partial t} = \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}}\right) N_2 + \frac{(N_2 - N_1)}{\tau_{21}} n_{ph} - \frac{N_1}{\tau_{10}}$$
(8)

$$\frac{\partial N_0}{\partial t} = \frac{N_1}{\tau_{10}} - \frac{N_0}{\tau_P} \tag{9}$$

$$N_{tot} = N_0 + N_1 + N_2 + N_3, (10)$$

where the population of the electronic energy level i is $N_i = N_i(\vec{r}, t)$, with $i \in \{0...3\}$ and N_{tot} is the total number of electrons participating in the lasing process. Furthermore, the photon number density is n_{ph} and defined as $n_{ph} = N_{ph}/N_{tot}$, it is driven by the population inversion in the so-called laser levels 1, 2. The electronic transition rates from level i to j are γ_{ij} and inverse to the respective life or relaxation times τ_{ij} , i.e. $\gamma_{ij} \equiv 1/\tau_{ij}$. Additionally, also non-radiative decay processes may occur with a rate of γ_{nr} . Considering the stationary limit, characterized by $\partial_t N_i = 0$, the above system of equations may be simplified. Assuming γ_{32} and γ_{10} large as compared to all other rates in the system, we find the population inversion between the upper and the lower laser level given as

$$n_2 = \frac{\gamma_P}{\gamma_P + \gamma_{nr} + \gamma_{21} \left(n_{ph} + 1\right)}.$$
(11)

2.2 Random Laser Model for a Finite System of ZnO Nano-Pillars

From the above discussions it becomes clear that uniformly pumped systems can only have a stationary lasing state, if the build up lasing intensity partly leaves the system, so that the dynamically generated gain, or population inversion, compensates for the losses out of the system. In the systems considered here this may occur in two different ways, the obvious way is through the system's boundaries and the other way is through leakage into the third dimension, i.e. along the direction of the nanopillars (the z-direction). The latter, is accounted for by an additional term in the propagating energy density or photon number density. In general, the light intensity n_{ph} as introduced above, obeys a diffusion equation,²⁸

$$\partial_t n_{ph} = D_0 \nabla^2 n_{ph} + \gamma_{21} (n_{ph} + 1) n_2, \tag{12}$$

where D_0 is the diffusion coefficient and $\nabla^2 = \partial_x^2 + \partial_y^2$. The last term, $\gamma_{21}(n_{ph} + 1)n_2$, describes the change in intensity due to photon emission, including both stimulated as well as spontaneous, cf. Eqs. (7), (8) and (11). As outlined and explained above we use the translational invariance in the x direction to perform a Fourier



Figure 3. Displayed is the geometry of the ZnO pillar system in the left panel. In the right panel we show the two dimensional setup as used in the actual calculations for the TM polarized electric field. The system is assumed to be translational invariant in the x direction and of finite width in the y direction. The system may experience light intensity loss through the surfaces due to this finite width or by leakage along the z direction. This leakage is considered as a free parameter in the calculation, to be adjusted to the experimental findings.

transform with respect to that coordinate. Eventually, we look for the energy response function $P^{\Omega}(Q_x, y)$, which is related to the photon density by

$$n_{ph}(\vec{r}) = \int_{-\infty}^{+\infty} \mathrm{d}^2 r' P^{\Omega}(\vec{r}, \vec{r}') n_2(\vec{r}').$$
(13)

By using the diffusion equation for the photon number, Eq. (12), in conjunction with the stationary state and the diffusive limit, the following expression is derived

$$P^{\Omega}(Q_x, y) = \frac{i\gamma_{21}}{\Omega - iQ_x^2 D_0 + i\chi^{-2} D_0 + i\zeta^{-2} D_0} , \qquad (14)$$

where we surpressed the relative coordinates and have derived the correlation length χ which represents the finite decay length of the intensity in this active medium and is defined as

$$\chi = \sqrt{\frac{D_0}{\gamma_{21}} \ \frac{n_{ph}}{n_2}} \ . \tag{15}$$

Furthermore, we introduced the last term in the denominator, $i\zeta^{-2}D_0$, by hand in order to model the additional loss due to leakage along the z direction in the model. The associated decay length ζ has to be extracted from experimental data. By defining an general decay length ξ for the light intensity response function, we can write

$$P^{\Omega}(Q_x, y) = \frac{i\gamma_{21}}{\Omega - iQ_x^2 D_0 + i\xi^{-2}D_0} , \qquad (16)$$

with

$$\frac{1}{\xi^2} = \frac{1}{\chi^2} + \frac{1}{\zeta^2} \tag{17}$$

This quantity has the dimension of length and describes, according to Eq. (14), how the intensity of the TM mode in the system in real space decays along the x direction for any given value of y within the nano-pillar system. This decay includes both, the intensity loss through the system's surfaces and gain through lasing action, as well as the leakage along the z-direction. We again point out that the according quantity ζ has to be inferred from experimental data.

Proc. of SPIE Vol. 8095 80950R-5



Figure 4. The intensity correlation length profile across the width W of the disordered nanopillar array as depicted in Fig. 3 for external pumping rates characterized by $\gamma_P/\gamma_{21} = 2$ and 4, i.e. above threshold. For the D_0 we used $D_0 = 0.01\gamma_{21}W^2$.

For a uniform pumping of this system, which is translational invariant in the x direction, the averaged light intensity does not depend on the x coordinate. Therefore, we have to consider the diffusion equation, cf. Eq. (12),

$$D_0 \partial_y^2 n_{ph}(y) = -\gamma_{21} (n_{ph}(y) + 1) n_2(y), \tag{18}$$

with the electronic population inversion n_2 given in the stationary state by

$$n_2(y) = \frac{\gamma_P}{\gamma_P + \gamma_{nr} + \gamma_{21} \left(n_{ph}(y) + 1 \right)}.$$
(19)

This results in the differential equation for the average photon number according to

$$\partial_y^2 n_{ph}(y) = -\frac{\gamma_{21}}{D_0} \frac{(\gamma_P / \gamma_{21})}{1 + \frac{(\gamma_P / \gamma_{21})}{n_{ph}(y) + 1}}.$$
(20)

The Equations (15), (19) and (20) constitute the solution for the intensity profile of TM polarized light in the disordered, lasing nano-pillar system. They also suffice to describe the correlation length defining the average volume over which the system typically collects enough intensity to cross the laser threshold and enters a stationary lasing mode.

3. RESULTS AND DISCUSSION

We study the behavior of disordered ZnO nanopillar arrays with a geometry as depicted in Fig. 3. In the left panel of Fig. 3, the displayed sketch gives an impression of the real system, which we assume to be translational invariant in the x direction and of limited width W in the y direction. The right panel in the same figure shows the actual setup as employed in the theory of TM polarized field diffusing effectively in 2d, encountering scattering events at circular scatterers.

3.1 Large Modes vs. Small Modes

As becomes clear from the previous section, there exist two fundamentally different ways of establishing a stationary lasing mode. One kind are *surface modes* that encounter in-plane intensity loss through the sample surfaces at $y = \pm 0.5W$ where W is the width, cf. Fig. 3. These modes have been observed as large modes in experiments,⁵ because they extent over the entire width of the sample. The other kind of lasing modes are pure *bulk modes*. These modes occur in regions with strong leakage in z direction, the direction of the pillars. Once the leakage is large enough to compensate for amplification by stimulated emission, a stationary lasing states is established, even without an intensity loss through the surfaces of the 2d system. The size of this mode then depends on the leakage as expressed by correlation length ζ , Eq. (15). In contrast to the surface modes, the bulk



Figure 5. Displayed is the calculated average area in which lasing intensity builds up, this corresponds to the size of the observed lasing spots. More specifically, it is the are over which the intensity decreases to 1/e of it its initial value. The geometry of the system is shown in Fig. 3, it is of limited width W in the y direction and translational invariant in the x direction. From left to right, in both panels, the pump rates are increasing $\gamma_P/\gamma_{21} = 1, 10, 20, 30, 40$, therefore, starting at the lasing threshold of the system. The difference between the system in the upper and the lower panel lies in the boundary conditions. In the upper panel, the laser intensity leakage through the surfaces located at $y/W = \pm 0.5$ is twice as much as in the lower panel. The two systems are otherwise identical, with $D_0 = 0.01\gamma_{21}W^2$. For a detailed discussion see text.

modes are more of circular shape and should typically be small compared to the width of the sample, because otherwise they would extent to surface modes, as we show below.

First we study the large or surface modes. We numerically evaluate Equations (15), (19) and (20) with symmetric boundary conditions, since the two bounding surfaces of the disordered nanopillar arrays are assumed



Figure 6. Displayed is the correlation length profile. The solid (black) line corresponds to Fig. 5, lower panel, left most plot. The dashed (red) curve represents the solution to a system with small intensity leakage into the z direction, but with otherwise unchanged parameters ($D_0 = 0.01\gamma_{21}W^2$). The additional and nondiffusive loss, $\zeta = 0.4W$, is reflected by the systems decreased area over which a stable lasing mode can exist.

to behave identical with respect to photon losses. By this, we obtain the spatial profile of all involved quantities, in particular also for the intensity correlation length ξ . A typical profile for external pumping rates characterized by $\gamma_P/\gamma_{21} = 2$ and $\gamma_P/\gamma_{21} = 4$, i.e. above threshold is shown in Fig. 4.

Characteristic features are that a stronger pumping rate reduces the correlation length and therefore, the area in which a stable lasing mode can exist, and the larger values of the correlation length deep inside the array and comparably smaller values at the surfaces, where there is not only a diffusive loss out of given mode but also strong intensity loss through the surfaces.

Results of a numerical evaluation are displayed in Fig. 5. There, in Fig. 5, we show the calculated average lasing spot sizes as calculated according to the above discussion. Since we assumed the system to be translational invariant in the x direction, we may choose any x point as a reference point, so we chose x = 0 as point of reference from which we show data. To be more specific, we show the area over which the intensity decreases to 1/e of it its initial value at the reference point (x, y) = (0, 0), also compare to the geometry as shown in Fig. 3. In Fig. 5 we consider two physical systems which are different in the photon loss rate through the surfaces, but are otherwise identical. In the upper panel, the homogeneous external pumping is increased, it assumes values of $\gamma_P/\gamma_{21} = 1, 10, 20, 30, 40$, where $\gamma_P/\gamma_{21} = 1$ marks the laser threshold because the equality between γ_P and γ_{21} characterizes the situation where electrons are as fast excited into the upper laser level as they relaxate to the lower laser level. We observe a decrease of the lasing area as the pumping increases, roughly speaking to 16%of its threshold value. As discussed, in the lower panel the boundary condition has been changed such that here the photon leakage rate through the surfaces located at $y/W = \pm 0.5$ is only half as much as in the upper panel. Therefore, more light intensity remains inside the array of nanorods and causes a severe reduction of the lasing spot size. This is to be interpreted such that the higher intensity inside the sample needs less area to establish a stable and stationary lasing mode. Finally in Fig. 6 we compare a surface mode with leakage into the direction of the pillars with the same surface mode without this kind of intensity loss. As seen in Fig. 6, the loss results in a decrease of the lasing spot size. This is different compared to the surface losses where an enlargement of loss increases the spot size. This additional loss, however, is nondiffusive in nature and reduces the intensity in a given area, therefore the stable lasing mode maintains its compensation equilibrium by reducing its area.

In order to discuss the bulk or small lasing modes, it is necessary to consider again Eq. (16). The additionally introduced decay length ζ , describing light intensity loss into the direction of the nanopillars, also affects the



Figure 7. Displayed is the calculated average area in which lasing intensity builds up, this corresponds to the size of the observed lasing spots. From right to left, we show a pure surface mode (the same as in Fig. 4 lower panel, second from left), in the middle a surface mode which also experiences intensity leakage into the direction of the nanopillars, and in the right panel we show a pure bulk mode, with the same leakage into the z direction but without contact to the system's boundaries. Otherwise, the system parameters are identical ($\gamma_P = 10\gamma_{21}, D_0 = 0.01\gamma_{21}W, \zeta = 0.4W$).

diffusing photon number. Taking this into account, and assuming a stationary state in the bulk with a homogeneous intensity distribution over its small size, we find the following condition that must be met by the photon number

$$\gamma_{21} \left(n_{ph} + 1 \right) n_2 = \frac{D_0}{\zeta^2} n_{ph}.$$
(21)

Therefore, for bulk modes we have to consider Eqs. (17), (19) and (21). In the following we numerically study such systems in detail. In Fig. 7, we directly compare the above bulk solution, in the right most panel, to the corresponding surface solutions. In particular to a perfect surface mode in the left panel, and to the same surface mode which additionally leaks intensity into the direction of the pillars. We observe the drastic change in the behavior of the lasing spot size, as was recently observed in experiments.⁵ The excitation of such a bulk mode, depends on two conditions which must be met, first there has to be a finite loss into the z direction, of course. Second, the physical system has to provide multiple scattering, gain and pump conditions, so that the laser threshold can locally be reached within a certain area, such that there is no contact to the actual surfaces of the 2d system. If this can be arranged, an equilibrium is established between light amplication within this coherence area and the loss along the z-direction. This compensation then enables a stable stationary lasing state without contact to the surfaces. In a spatially disordered system with strong enough scattering to cause diffusive light transport, the fullfillment of these conditions appears realistic, given that the sample width remains large enough, to justify true two dimensional behavior. In very small, i.e. thin, systems, only surface modes are expected to be present.

4. CONCLUSIONS

In conclusion, we have derived and discussed an analytical diffusion theory for random lasing in effectively two dimensional disordered systems of nanopillars of finite spatial size. The included intensity losses through the sample's surfaces together with a possible leakage along the direction of the rods enable the existence of stable lasing mode, in which a stationary compensation between losses and light amplification is established. Our theory allows us to compute the intensity correlation length which corresponds to the observed average lasing spot size. The presented study of the behavior of this lasing spot size also include a free parameter to describe the average leakage along the pillar direction. This additional leakage introduces to fundamentally different lasing modes. The large surface mode, extending over the entire width of the sample, and the small bulk mode, without contact to the surfaces. The bulkmode therefore emits into he leakage direction along the nanopillars. Our new findings of two different types of lasing modes in disordered 2 dimensional systems agrees with experimental data.⁵ Furthermore, for the two dimensional surface modes we predict a decrease of the correlation length, and therefore accordingly for the spot size, which is proportional to $\xi \propto 1/\sqrt{\gamma_P}$. Bulk modes behave differently due to their constant loss.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge helpful discussions with D. Chigrin, J. Kroha, H. Kalt, A. Lavrinenko, U. Lemmer, G. Schön, B. van Tiggelen, D. Wiersma. Furthermore we acknowledge support by the Karlsruhe School of Optics & Photonics (KSOP) (R.F.) and the SFB 608 (A.L.).

REFERENCES

- V. S. Letokhov, "Generation of light by a scattering medium with negative resonance absorption.", Sov. Phys. JETP 26, 835 (1968).
- [2] H. Cao, Y. G. Zhao, H. C. Ong, S. T. Ho, J. Y. Dai, J. Y. Wu, R. P. H. Chang, "Ultraviolet lasing in resonators formed by scattering in semiconductor polycrystalline films.", *Appl. Phys. Lett.* 73, 3656 (1998).
- [3] H. Cao, Y. G. Zhao, S. T. Ho, E. W. Seelig, Q. H. Wang, R. P. H. Chang, "Random Laser Action in Semiconductor Powder.", *Phys. Rev. Lett.* 82, 2278 (1999).
- [4] V. M. Markushev, V. F. Zolin, C. M. Briskina, "Powder laser.", Zh. Prikl. Spektrosk. 45, 847 (1986).
- [5] J. Fallert, R. J. B. Dietz, J. Sartor, D. Schneider, C. Klingshirn, H. Kalt, "Co-existence of strongly and weakly localized random laser modes.", *Nature Photonics* 3, 279 (2009).
- [6] K. L. van der Molen, P. Zijlstra, A. Lagendijk, A. P. Mosk, "Laser threshold of Mie resonances.", Opt. Lett. 31, 1432 (2006).
- [7] M. Bahoura, K. J. Morris, M. A. Noginov, "Threshold and slope efficiency of Nd_{0.5}La_{0.5}Al₃(BO₃)₄ ceramic random laser: effect of the pumped spot size.", Opt. Commun. 201, 405 (2002).
- [8] N. M. Lawandy, R. M. Balachandran, A. S. L. Gomes, E. Sauvain, "Laser action in strongly scattering media.", *Nature* 368, 436 (1994).
- [9] S. V. Frolov, Z. V. Vardeny, K. Yoshino, A. Zhakidov, R. H. Baughman "Stimulated emission in high-gain organic media.", *Phys. Rev. B* 59, R5284 (1999).
- [10] S. Gottardo, S. Cavalieri, O. Yaroshchuk, D. A. Wiersma "Quasi-two-dimensional diffusive random laser action.", *Phys. Rev. Lett.* 93, 263901 (2004).
- [11] R. C. Polson, Z. V. Vardeny, "Random lasing in human tissues.", Appl. Phys. Lett. 85, 1289 (2004).
- [12] S. Klein, O. Cregut, D. Gindre, A. Boeglin, K. D. Dorkenoo, "Random laser action in organic film during the photopolymerization process.", *Opt. Express* 3, 5387(2005).
- [13] M. Anni, S. Lattante, T. Stomeo, R. Cingolani, G. Gigli, G. Barbarella, L. Favaretto, "Modes interaction and light transport in bidimensional organic random lasers in the weak scattering limit.", *Phys. Rev. B* 70, 195216 (2004).
- [14] F. Quochi, F. Cordella, R. Orrú, J. E. Communal, P. Verzeroli, A. Mura, G. Bongiovanni, "Random laser action in self-organized para-sexiphenyl nanofibers grown by hot-wall epitaxy.", *Appl. Phys. Lett.* 84, 4454 (2004).
- [15] H. Cao, "Lasing in random media.", Waves Random Media 13, R1 (2003).
- [16] H. Cao, "Review on latest developments in random lasers with coherent feedback.", J Phys A: Math General 38, 10497 (2005).
- [17] H. Cao, J. Y. Xu, D.Z Zhang, S. H. Chang, S. T. Ho, E. W. Seelig, X. Liu, R. P. H. Chang, "Spatial Confinement of Laser Light in Active Random Media.", *Phys. Rev. Lett.* 84, 5584 (2000).
- [18] H. Cao, Y. Ling, J. Y. Xu, C. Q. Cao, P. Kumar, "Photon statistics of random lasers with resonant feedback.", *Phys. Rev. Lett.* 86 4524 (2001).
- [19] C. W. J. Beenakker, "Photon statistics of a random laser.", Diffusive waves in complex media 86 4524 (1999).
- [20] V. M. Apalkov, M. E. Raikh, B. Shapiro, "Random resonators and prelocalized modes in disordered dielectric films.", *Phys. Rev. Lett.* bf 89, 016802 (2002).

- [21] C. Vanneste, P. Sebbah, H. Cao "Lasing with resonant feedback in weakly scattering random systems.", *Phys. Rev. Lett.* bf 89, 143902 (2007).
- [22] P. W. Anderson, "Absence of Diffusion in Certain Random Lattices.", Phys. Rev. 109, 1492 (1958).
- [23] E. Abrahams, P. W. Anderson, D. C. Licciardello, T. V. Ramakrishnan, "Scaling Theory of Localization: Absence of Quantum Diffusion in Two Dimensions.", *Phys. Rev. Lett.* 42, 673 (1979).
- [24] D. Vollhardt and P. Wölfle, "Diagrammatic, self-consistent treatment of the Anderson localization problem in $d \le 2$ dimensions.", *Phys. Rev. B* 22, 4666 (1980).
- [25] R. Frank, A. Lubatsch, J. Kroha, "Theory of strong localization effects in disordered loss or gain media.", *Phys. Rev. B* 73, 245107 (2006).
- [26] R. Frank, A. Lubatsch, "Scalar wave propagation in random amplifying media: Influence of localization effects on length and time scales and threshold behavior.", *Phys. Rev. A* 84, 013814 (2011).
- [27] R. Frank, A. Lubatsch, J. Kroha, "Light transport and localization in diffusive random lasers.", J. Opt. A: Pure Appl. Opt. 11, 114012 (2009).
- [28] L. Florescu, S. John, "Lasing in a random amplifying medium: Spatiotemporal characteristics and nonadiabatic atomic dynamics.", *Phys. Rev. E* 70, 036607 (2004).
- [29] X. Y. Jiang, C. M. Soukoulis, "Time dependent theory for random lasers.", Phys. Rev. Lett. 85, 70 (2000).
- [30] A. Yamilov, S. H. Chang, A. Burin, A. Taflove, H. Cao, "Effects of localization and amplification on intensity distribution of light transmitted through random media.", *Phys. Rev. E* 70, 037603 (2004).
- [31] S. H. Chang, A. Taflove, A. Yamilov, A. Burin, H. Cao, "Numerical study of light correlations in a random medium close to the Anderson localization threshold.", *Optics Lett.* **29**, 917 (2004).
- [32] A. L. Burin, M. A. Ratner, H. Cao, R. P. H. Chang, "Model for a random laser.", Phys. Rev. Lett. 87, 215503 (2001).
- [33] A. Yamilov, H. Cao, "Field and intensity correlations in amplifying random media.", Phys. Rev. B 71, 092201 (2005).
- [34] J. Fallert, R. J. B. Dietz, H. Zhou, J. Sartor, C. Klingshirn, H. Kalt, "Lasing in single ZnO nanorods after fs- and ns-pulsed excitation.", phys. stat. sol. (c) 6, 449 (2009).
- [35] Y. Li, M. Feneberg, A. Reiser, M. Schirra, R. Enchelmaier, A. Ladenburger, A. Langlois, R. Sauer, K. Thonke, J. Cai, and H. Rauscher, "Au-catalyzed growth processes and luminescence properties of ZnO nanopillars on Si.", J. Appl. Phys. 99, 054307 (2006).
- [36] S. Mandal, K. Sambasivarao, A. Dhar, and S. K. Ray, "Photoluminescence and electrical transport characteristics of ZnO nanorods grown by vapor-solid technique.", J. Appl. Phys. 106, 024103 (2009).