Light Propagation in Disordered Two Dimensional Media and Random Lasing Modes

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Abstract. We present a theory for light propagation and light intensity transport in thin random dielectric media with optical gain. The diffusing light intensity is coupled to the semiclassical laser rate equations. This coupled system of nonlinear equations is numerically solved for the stationary laser states. Our calculations predict two different types of random laser modes in effectively two-dimensional systems. Surface modes which exhibit a large size and extend over the entire sample width, and also bulk modes with comparably small lasing areas.

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

Transport in random media is on the threshold to gain significant technological importance in a broad field of physics reaching from transport in graphene on the one hand side up to light management in solar cell science. Enhancing the efficiency of solar cells by comparable cheap techniques which may be described by 'light management from the aerosol can' is within reach and disordered scatterers are considered to improve light harvesting in tailored solar cells. Another important aspect of transport theory is the random laser theory based on a field-theoretical ansate which combines both, transport of light and localization in disordered media and microscopic amplification and stimulated emission is a most intriguing theoretical system. Even in the absence of a resonator, stimulated emission and lasing action occurs as was first pointed out already in 1968 [1]. On the experimental side physical systems ranging from powders of semiconductor nanoparticles [2, 3, 4, 5, 6, 7, 8], to ceramics [9], to organic laser dyes placed in strongly scattering media [10, 11, 12], to organic films or nanofibers [13, 14, 15, 16], are experiencing a spiraling growth of interest.

LIGHT TRANSPORT IN DISORDERED MONOLAYERS OF MIE SPHERES

The here considered system is schematically depicted in Fig. 1 and consists of small spherical scatterers, which are themselves composed of a homogeneous medium, such as *ZnO*. This material is in particular laser active, therefore offering the possibility to observe lasing action. The scatterers in our model are assumed to be identical and located at random positions. We consider very thin films up to monolayers, requiring a two dimensional treatment. The host medium surrounding the spherical scatterers may any dielectric including absorption and optical gain. This system is practically uniformly pumped, depicted be the red light cone in Fig. 1. A strong enough external pumping of the laser active medium in the scatterers leads to the observation of lasing emission from certain, well localized, spatial areas in the sample. This is schematically indicated as the bright magenta spot in Fig. 1. These lasing spots occur at random position, with a mean size depending on system parameters, such as scatterer mean size and their density per volume, pump strength and intensity leakage to the environment.

We study the behavior of a positionally disordered arrangement of spherical *ZnO* scatterers with a geometry as depicted in Fig. 1. We assume a translational invariance in the x direction and a limited width W in the y direction. In an optically active system, stationary lasing modes can only exist if there is also intensity loss out of the system. These losses are only to be provided by the finite system size, i.e. by surfaces.

The lasing behavior can be described in terms of semiclassical rate equations for the occupation numbers of the involved electronic energy levels of the underlying active material, as given by

$$\frac{\partial N_3}{\partial t} = \frac{N_0}{\tau_P} - \frac{N_3}{\tau_{32}} \quad (1)$$

$$\frac{\partial N_2}{\partial t} = \frac{N_3}{\tau_{32}} - \left(\frac{1}{\tau_{21}} + \frac{1}{\tau_{nr}}\right) N_2 - \frac{(N_2 - N_1)}{\tau_{21}} n_{ph} \quad (2)$$

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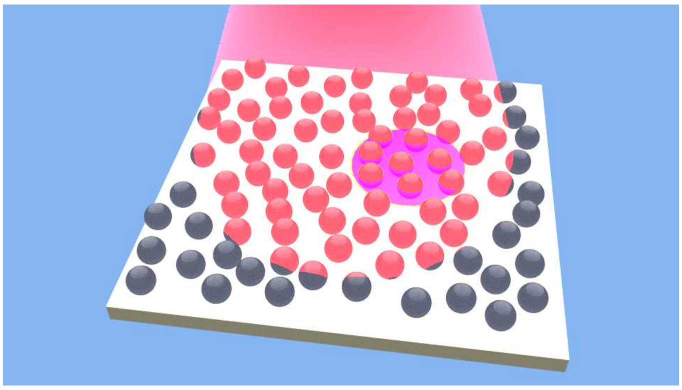

FIGURE 1. The described system consists of small spherical scatterers, which are themselves composed of a homogeneous medium, such as *ZnO*. This material, in particular, is laser active. The scatterers in our model are assumed to be identical and located at random positions. We consider very thin films up to monolayers, requiring a two dimensional treatment. The host medium surrounding the spherical scatterers may any dielectric including absorption and optical gain. This system is practically uniformly pumped, depicted be the red light cone. Strong enough pumping leads to the observation of lasing emission from certain regions in the sample. 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 scatterer size and density per volume, pump strength and intensity leakage to the environment.

$$\frac{\partial N_1}{\partial t} = \left(\frac{1}{\tau_{21}}+\frac{1}{\tau_{nr}}\right)N_2+\frac{(N_2-N_1)}{\tau_{21}}n_{ph}-\frac{N_1}{\tau_{10}} \quad (3)$$

$$\frac{\partial N_0}{\partial t} = \frac{N_1}{\tau_{10}}-\frac{N_0}{\tau_P} \quad (4)$$

$$N_{tot} = N_0+N_1+N_2+N_3. \quad (5)$$

In the above equations, we introduced the electronic occupation numbers N_i of level i, relaxation times from level i to j as τ_{ij}, the photon number density n_{ph} of the lasing mode and the pumping in terms of τ_p.

These rate equations are to be completed by a transport model for the photon number density transport within the random, active medium. This intensity propagation obeys a diffusion equation accounting for both the multiple scattering as well as optical amplification due to stimulated emission, which reads [28],

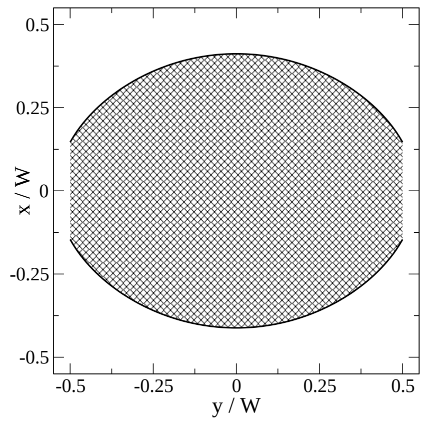

FIGURE 2. Calculated average area of a *surface mode* corresponding to the size of the observed lasing spots. The system parameters are $\gamma_P = 2\gamma_{21}, D_0 = 0.2\gamma_{21}W, \zeta = 0.0W$.

$$\partial_t n_{ph} = D_0\nabla^2 n_{ph}+\gamma_{21}(n_{ph}+1)n_2, \quad (6)$$

where n_{ph} is the photon number density and furthermore, D_0 is the diffusion coefficient and $\nabla^2 = \partial_x^2+\partial_y^2$. The last term on the right-hand-side of Eq. (6), i.e. $\gamma_{21}(n_{ph}+1)n_2$, describes the intensity gain due to stimulated photon emission, cf. Eqs. (2), (3). In order to account for the out-of-plane losses of lasing intensity, we have to further introduce an additional term in the above diffusion equation, Eq. (6). Therefore, we find eventually

$$\partial_t n_{ph} = D_0\nabla^2 n_{ph}+\gamma_{21}(n_{ph}+1)n_2-\frac{D_0}{\zeta^2}n_{ph}, \quad (7)$$

where ζ characterizes the intensity leakage.

We are in particular interested in the stable, stationary laser modes. Therefore, we numerically solve Eqs. (1) to (5) and Eq. (7) incorporating suitable boundary conditions at the system surfaces, for the stationary states only. The calculated results for the light intensity correlation length or spot size are displayed in Fig. 2 and Fig. 3, for the surface and the bulk mode, repsectively.

DISCUSSION AND CONCLUSION

In conclusion, we discuss a diffusion theory for random laser systems of finite size in effectively two spatial dimensions for disordered systems of spherical scatters. We consider both, light intensity losses in-plane as well as out of plane. The included in-plane losses are through the sample's surfaces, the out-of-plane intensity leakage is along the direction perpendicular to plane. Both of

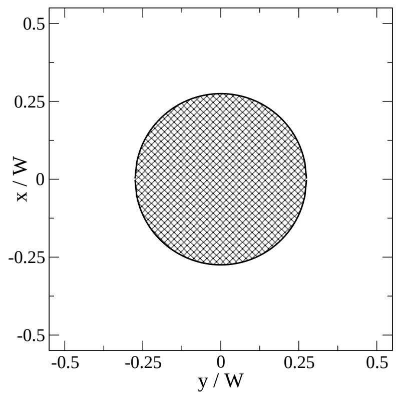

FIGURE 3. Calculated average area of a *bulk mode* in which lasing intensity builds up, i.e. the spot size. The system parameters are $\gamma_P = 2\gamma_{21}, D_0 = 0.2\gamma_{21}W, \zeta = 0.6W$.

these two loss mechanisms are the reason for the existence of stable lasing modes, in which a stationary compensation between losses and light amplification is established. The developed theory allows for the computation of the average size of lasing spots, which correspond to intensity correlation length. The possibility of out-of-plane leakage introduces two lasing modes, which differ through their loss mechanisms. The large surface mode Fig. 2 , extending over the entire width of the sample, and the small bulk or volume mode Fig. 3, without contact to the surfaces. The bulk mode therefore emits into he leakage direction perpendicular to the system plane. The finding of two different types of lasing modes in disordered two dimensional systems agrees with recent experimental data [7]. For the two dimensional surface modes we predict a decrease of the intensity correlation length with increasing pump strength, proportional to $\xi \propto 1/\sqrt{\gamma_P}$. Bulk modes behave differently.

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