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Physica B 338 (2003) 212–214
PHYSICA B
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Understanding and control of random lasing ☆

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Abstract

Random lasing attracts much attention because it helps to understand coherent phenomena in disordered media and can be used in optoelectronics due to easy preparation (no need in mirrors) and small size of random lasers down to few microns. Recently the remarkable progress in studying the material, geometry and external pumping dependences of laser properties and efficiency has been reached. Lasing emerges from the special random cavities of high quality formed within the active medium. They can be described as the decaying eigenoptical modes within the medium and the optical mode having the minimum decay rate is responsible for lasing. Numerical and analytical studies of the properties of these modes permit to interpret existing experiments and suggest the ways to optimize the performance of lasers.

PACS: 71.55.Jv; 42.25.Fx; 42.55.−f

Keywords: Random laser; Open system; Nanoparticles; Cavity

Random lasers are microsize mirror less sources of coherent light interesting both from fundamental and applied points of view [1]. The significant practical problem here is the reduction of lasing threshold, i.e., increase of the quality of medium modes, responsible for lasing [2]. The abrupt change of optical properties of medium takes place near the lasing threshold, where the only one mode with the longest lifetime dominates over others, as opposed to the passive medium. To enable the efficient control of lasing we need to understand the parametric dependence of lasing properties (our analysis in this brief paper is restricted to the lasing threshold). Clarifying measurements [3] of the lasing threshold dependence on the light transport length l_t, the pump area and the dye concentration n_{dye}, performed in the lasing medium composed by rhodamine dye (for lasing emission) and TiO_2 nanoparticles (to provide controllable scattering) dissolved in PMMA can help to understand better lasing from 3D random medium (analysis of lasing in open geometry ZnO [3], where lasing has been discovered [4], and of ordering and localization effects

☆ This work was supported by the DoD MURI program, by the MRSEC/NSF program through the Northwestern University MRSEC (grant no. DMR-00076797), and partially by the NSF (Grant No. ECS-9877113). H.C. acknowledges support from David and Lucile Packard and Alfred P. Sloan Foundations. AB acknowledges Mikhael Noginov for fruitful discussions.

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doi:10.1016/j.physb.2003.08.031

has been performed in our previous works [2]). We assume the system to be far from Anderson localization regime [3].

The experiment has demonstrated that the lasing threshold intensity I_{thr} (minimum pump intensity needed to reach lasing) decreases with the light transport length l_t ($I \propto l_t^{1/2}$ [3]), with the beam diameter d ($I \propto 1/d^a$, $1 \leqslant a \leqslant 2$, [3,5] saturating at $d \sim 200$ μm [3] or 1 mm [5]). It also decreases with the increase of the dye concentration [3]. If we assume that the gain coefficient g in the active medium is proportional to the absorbed pump power within the cylinder domain of the depth $l_p \sim \sqrt{l_{t1} l_{abs}/3} < 10$ μm (here l_{t1} is the transport length of the pump light and $l_{abs} \sim 6$ μm is its absorption length), and the diameter $d \gg l_p$ [3], then according to the diffusion model [6] the lasing threshold should not depend on the beam spot diameter d under experimental conditions $d \gg l_p$ where it is defined entirely by the gain cylinder height

$$\frac{1}{l_g} \sim \frac{l_t}{l_p^2}, \tag{1}$$

where $1/l_g$ is the gain length. To explain the deviations from the diffusion behavior the model of strong fluctuations has been suggested in Ref. [3]. It was expected that the lasing mode having highest quality is very different from the typical diffusion solution and have longer lifetime strongly sensitive to this anomalous mode geometry. This model makes some sense for the open system (planar layer of ZnO [2]), while the analysis of the 3D mode life-time statistics within the random matrix approach [7] basically confirms the diffusion model expectations. To verify whether the fluctuations break down the diffusion model predictions we performed the numerical analysis of 2D closed system of scattering particles (infinite height cylinders) within the resonant approach suggested in Ref. [3], generalized to 2D geometry. We found that until the 2D localization shows up (at very large sample size) the inverse dependence on the squared sample length (1) predicted by the diffusion model takes place [8]. The analysis in more detail will be published elsewhere. These results agree with the recent preprint [9]. The fluctuations in 2D should be much stronger than those in 3D, so we do expect that the diffusion model should work in 3D even better.

Thus the experiment [3] should be explained within the diffusion model. We believe that the seeming contradiction between experiment and theory is caused by the saturation in the absorption, which has been ignored in Ref. [3]. One can expect that the same dye molecule is not able to absorb more than one photon during the 25 ps pumping pulse used in Ref. [2]. We estimate the ratio of the number of incoming photons and dye molecules within the gain cylinder for the typical parameters including [3] lasing threshold intensity $I \sim 2$ MW/mm^2, dye concentration $n_d \sim 0.02$ M, transport length $l_{t1} \sim 1$ μm (leading to the penetration length $l_p \sim 1.4$ μm, below we assume that the transport lengths for the pumping l_{t1} and emitted light l_t coincide, or they are at least proportional to each other) to be about 10. Thus even if we assume (ignoring for instance the inhomogeneous broadening of absorption spectrum) that each dye molecule can absorb one photon, other photons will go essentially deeper.

To describe the saturated absorption consider the following simple model. There is the maximum possible absorbed intensity I_s and the absorption coefficient depends on the intensity of pump light as $\alpha \equiv \alpha_0 I_s/(I + I_s)$ (the linear absorption length l_{abs} and the linear absorption coefficient are related as $1/l_{abs} = \alpha_0/v$, where v is the speed of light in the medium). Assume that the pumping intensity I_0 is much larger than the limiting value I_s. Then the pump light will go inside the medium much deeper than the linear penetration length $l_p \sim 1$ μm to some new characteristic depth R. If it exceeds the beam diameter $R > d$ then the incoming intensity is absorbed within the volume R^3 and the total absorbed power can be estimated as $\alpha_0 I_s R^3$ ($\alpha_0 I_s R d^2$ for $R < d$). The gain length within the gain volume R^3 is independent on the incoming intensity and corresponds to its absolute possible minimum l_{gm}, which depends on the dye concentration n_{dye} only ($1/l_{gm} \propto n_{dye}$).

To find the gain volume size R one can use the energy conservation condition. The absorbed energy should be equal to the incoming flux $j \sim D \nabla I d^2$. In the case of the large pump area $d > R$ one can set $\nabla I \sim I/R$ and the energy balance

reads $j \sim v l_t I_0 d^2/R \sim v I_s R d^2/l_{abs}$, where I_0 is the pump intensity. In the opposite case of small area the typical value of the derivative is $\nabla I \sim I/d$ because the significant decrease of intensity occurs on the size of the beam spot. If we neglect the outgoing flux at distances longer than d from the pump spot center then the energy balance can be written as $j \sim v l_t I_0 d^2/d \sim v I_s R^3/l_{abs}$. It is not generally quite right to ignore outgoing flux. We performed the numerical study of the saturated absorption that demonstrates the approximate validity of our arguments when d is smaller but comparable with R ($d > R/5$). This regime corresponds to the experimental situation [3], so that our arguments can be used to interpret the experiment.

According to the diffusion model [6], Eq. (1) the lasing instability takes place when $l_t/R^2 \sim 1/l_{gm}$. In the case of small pump beam diameter $d < R \sim (l l_{gm})^{1/2}$ the diffusion model (together with the energy conservation arguments above) yields the threshold intensity $I_{thr} \sim l_t^{1/2} l_{gm}^{3/2}/(l_{abs} d) I_s$, while at large d it changes to $I_{thr} \sim I_s l_{gm}/l_{abs}$. The first limit reproduces the experimental observations including $I_{thr} \propto \sqrt{l_t}$ dependence on the transport length [3] and inverse proportionality of the lasing threshold to the beam diameter [3,5]. At larger d both dependences are expected to be saturated. This seems to agree with Ref. [3] and the typical value of minimum gain length can be estimated in the mm scale that is reasonable. To analyze the I_{thr} dependence on the dye concentration n_{dye} we assume that $l_{gm} \propto 1/n_{dye}$, $I_s \propto n_{dye}^0$ and the linear absorption length $l_{abs} \propto 1/n_{dye}$. Then we find that for the low dye concentration, where $1/l_{gm} < l/d^2$ I_{thr} decreases with increasing n_{dye} as $n_{dye}^{-1/2}$ while at larger concentration it reaches the universal minimum $I_{min} = I_{sat} l_{gm}/l_{abs}$ that does not depend on the transport length, beam spot properties and dye density. The obtained dependence on the dye concentration agrees qualitatively with the experiment [3]. Note that even the minimum lasing threshold seems to exceed the saturation value of pump intensity because the gain length is larger than the absorption length. Therefore, one would expect lasing in 3D random medium to be always in the saturated absorption regime for pump light.

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