Model for a Random Laser

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The laser action in random media is studied numerically for a planar system of resonant scatterers pumped by an external laser. The eigenmodes of the finite system (quasistates) are "lossy" in the absence of gain because of the leakage of light outside the medium and can be characterized by their decay rates. Lasing occurs when the gain compensates the decay rate of the quasistate with the longest lifetime. The dependence of the lasing threshold on the number of scatterers (size of the system) is found to be $I \propto 1/\sqrt{N}$, which agrees with recent experiments. We demonstrate that this dependence is strongly related to the fluctuations of quasistate decay rates and discuss the nature of these fluctuations.

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The lasing instability in random media was predicted theoretically by Letokhov [1] more than 30 years ago. Remarkable progress in the experimental study of this phenomenon has been achieved in the past decade with the discovery of the amplified spontaneous emission [2] and the lasing action [3]. Random lasing has been found in various materials including ZnO powder [3,4], solution of TiO_2 nanoparticles, and rhodamine dye in polymethylmethacrylate (PMMA) [4,5] and in several polymer systems [6]. Random lasing with nonresonant feedback is seen as the remarkable narrowing of the luminescence spectrum to a single peak of width about several nanometers, while the coherent feedback lasing is identified as the series of high and narrow peaks having width decreasing with increase of the pump power to at least the tenth nanometer scale. Recent photocounting measurements [7] have proved that these peaks possess the Poisson photon statistics typical for a standard laser [8].

The random laser uses a microsize mirrorless cavity and can be very efficient in nanotechnology applications [9]. Two different theoretical approaches to random lasing can be distinguished. The analysis of average system response performed within the diffusion model possibly including coherent backscattering corrections [1,2,10,11] seems to be appropriate for describing the regime of nonresonant feedback (see, e.g., [10]), but it fails to predict the lasing threshold behavior for the laser action [3,5]. We think that the diffusion approach ignores rare fluctuations leading to the formation of high quality random cavities responsible for lasing and the right method should be based on a microscopic approach. Existing studies of one-dimensional systems [12,13] are affected by Anderson localization absent in real lasers [3–5]. A random matrix based analysis of the "lossy" chaotic 3D system has been suggested in Refs. [14]. It is not clear whether the random matrix approach can be applied to rare fluctuations in the resonant modes leading to random lasing. The comparison of our study and Ref. [14] will be made below and some problems will be discussed.

The goal of this paper is to study random lasing in a first principles model based on experiment. The medium is represented by a set of random scatterers treated in the resonant approach with the gain inside. We consider a planar two-dimensional medium that models the ZnO disk-shaped powder samples having relatively low lasing thresholds [3,5]. Even in that very lossy system the lasing threshold is very sensitive to the geometry and can be changed by orders of magnitude by increasing the size of the ZnO powder disk. This size dependence is reproduced in our modeling study, and most attention is paid to the origin of this dependence in terms of the properties of random high quality cavities, responsible for the lasing. In fact, the random laser efficiency is controlled by the properties of material and geometry. It is clear that ZnO particles provide a very efficient choice of the material while the optimum laser geometry is in question. We hope that our study would be useful in resolving that question.

Consider the optical excitations within a medium composed of N particles. General analysis is very complicated but it can be simplified if we replace all particles by dipolar oscillators. This approach is qualitatively valid for Mie resonances [15] within dielectric particles and for the surface plasmons within metal nanoparticles [16], and it is used in the majority of theoretical studies. Each particle k is considered as a point oscillator with the resonant frequency ω_k and transition dipole moment $\mathbf{d}_k$ similar to resonant atoms. The collective eigenmode can be characterized by the complex frequency z. The motion equation for a single oscillator k polarization component $\mathbf{p}_k = p_k\mathbf{d}_k/d_k$ belonging to this mode in the electric field $\mathbf{E}_k$ of other particles reads (we set $\hbar = 1$)

$$-z^2 p_k = -(\omega_k - ig)^2 p_k + 2\omega_k d_k(\mathbf{d}_k\mathbf{E}_k). \quad (1)$$

The gain is introduced into each scatterer by adding an imaginary term ig to its resonant frequency. This corresponds to pumping, when the external light is absorbed by the resonant scatterers, as for ZnO spheres [3,5].

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The local electric field $\mathbf{E}_k$ at a given frequency z and a wave vector $q = z/c$ in the system of N dipoles can be defined as the superposition of contributions found from the solution of the Maxwell equations for a single dipole electric field

$$\begin{aligned} \mathbf{E}_k &= \sum_{j \neq k} \mathbf{E}_{kj} + i\,\frac{2}{3}\,q^3 \mathbf{p}_k\,, \\ \mathbf{E}_{kj} &= e^{iqR_{kj}} \frac{\mathbf{p}_j - 3\mathbf{n}(\mathbf{n}\mathbf{p}_j)}{R_{kj}^3} (1 - iqR_{kj}) \\ &\quad + q^2 e^{iqR_{kj}} \frac{\mathbf{p}_j - \mathbf{n}(\mathbf{n}\mathbf{p}_j)}{R_{kj}}; \qquad \mathbf{n} = \frac{\mathbf{R}_{kj}}{R_{kj}}\,, \end{aligned} \tag{2}$$

where $\mathbf{R}_{kj}$ is the vector between two scattering centers k and j. The imaginary parts of contributions to the electric field are responsible for the light leakage that has to be overcome by the gain to get lasing. They have the standard form of spontaneous emission rates for particles treated similarly to resonant atoms. We consider this and only this part of the total leakage dependence on the specifics of the collective modes of light inside the medium [17] because they are sensitive to the interference.

Equations (1) and (2) form a linear system of N equations. It has N different solutions characterized by the collective eigenfrequencies $z_\alpha = \omega_\alpha + i\gamma_\alpha$, where γ_α is the decay rate due to the light emission outside the system. In the absence of the gain [$g = 0$, Eq. (1)] all decay rates are greater than 0 and the system is lossy. Increase of g leads to decreasing of γ_α. At some finite $g = g_*$ the decay rate for some mode vanishes. This indicates the occurrence of the lasing instability since the stationary response to any internal source becomes infinite for $g > g_*$.

When the resonant frequences of scatterers are close to each other the iteration method can be applied to find the lasing threshold. We start with some real input frequency ω_0, defined as the average resonant frequency of scatterers and compute the right hand side of the system Eqs. (2) and (1). Keeping the coupling terms at this fixed frequency we compute the set of the collective eigenfrequencies z_α in the left hand side. Then the complex frequency $z_\beta = \omega_\beta + i\gamma_\beta$ with the lowest decay rate γ_β is chosen as the output. At the next step we take the new input frequency ω_β and introduce the gain $g = \gamma_\beta$. The procedure is repeated until we get a real eigenfrequency ω_* at the gain g_*. This limiting value of the gain defines the lasing threshold. The convergence is fast and the result does not depend on the starting point. To rule out the nonphysical effect of very close pairs of particles we restrict our analysis to ω_* deviating from ω_0 by less than 20% and consider the effect of finite minimum distance $r_{\min}$ between particles up to the value of 0.4 of the average distance for $N \sim 100$ (at higher $r_{\min}$ the generation of random particle ensemble is complicated due to the large filling factor). Although the increase of $r_{\min}$ increases the lasing threshold, the overall increase at highest $r_{\min}$ does not exceed 30% of its value at $r_{\min} = 0$ and the parametric dependence does not change much compared to that case. Therefore in our analysis we use the results for $r_{\min} = 0$. Since the lasing threshold depends on the sample, we have computed its average value $g(N)$ and the standard deviation $\delta(N)$. Averaging is made over about 100 samples; this gives sufficient accuracy. The simulations were performed using OCTAVE [18] software.

Our simulations were proposed to approach the experimental conditions and to reveal the most significant parameters, affecting the lasing threshold. Because of computational difficulties we cannot treat a very large system with the number of particles exceeding $N \sim 1000$. Therefore the analysis within this letter is restricted to a two dimensional system of scatterers similar to the ZnO film of ~300 nm thickness on the sapphire substrate described in Refs. [3,5,19]. Since the typical size of the particle ~100 nm is close to ZnO disk thickness we can treat the sample as planar. The mismatch of dielectric constants of sapphire and air can affect the definition of the electric field (2), but it should not change results qualitatively. Therefore we ignore it. We maintain constant particle density, and investigate the behavior as system size (particle number) changes.

The sample is pumped by external laser of high frequency and the incident intensity is absorbed directly by the ZnO particles. Therefore the gain occurs inside the particles. The resonant mode inside each ZnO particle can be formed, for instance, by Mie resonance. The measured lasing threshold turns out to be very sensitive to the area of the ZnO planar disk (number of particles). This can be explained by the formation of high quality collective modes inside the layer. Since the collective excitations can be formed most efficiently when the particles are in resonance with each other, the case of all the same resonant frequencies $\omega_k = \omega_0$ [Eq. (1)] has been studied. In this case we expect the strongest sensitivity to the geometry. The positions of all N particles are generated randomly within the circle of the radius $R = \sqrt{N}\,\eta\,\lambda\!\!\!^{-}$, where $\lambda\!\!\!^{-} = c/\omega_0 = 1/q$ is the resonant wavelength. We know that experimentally $\eta \sim 1$ for the ZnO powder but cannot define its actual value. Therefore three different values of the density determining parameter $\eta = 0.3, 1, 3$ have been probed. Although in some cases (especially for $\eta = 0.3$) the interpoint distance in the random sample can be less than the minimum interparticle separation given by the particle diameter $\sim\lambda\!\!\!^{-}$, the effective minimum radius, defining the interaction, can be smaller than the separation of centers because of the specifics of the dipole-dipole interaction [16]. Simulations in which configurations corresponding to physically overlapping particles were excluded gave results almost identical to those with random configurations. In addition, our modeling study for the case $\eta = 0.3$ is intended to examine the effect of a relationship between the interparticle distance and the resonant wavelength rather than to reproduce explicitly the experimental conditions (this remains the difficult problem). The transition dipole moment has been taken with the same absolute values and random direction for each particle. One should note that our approach is crude since experimentally the particles are different, but we hope that the collective mode properties have some degree of universality and therefore can be described qualitatively by our approximation.

The results of simulations are shown in Fig. 1. The effect of collective modes (geometry) is actually strong since the lasing threshold changes by almost 2 orders of magnitude with increasing the particle number. The lasing threshold g decreases with the number of particles as $g \propto N^{-\alpha}$ that is equivalent to $1/A^{\alpha}$ dependence on the sample area at fixed density. Optimum fit yields $\alpha = 0.51$ for $\eta = 1$, $\alpha = 0.35$ for $\eta = 3$, and $\alpha = 0.52$ for $\eta = 0.3$. Experimental study of the two-dimensional ZnO powder [5] shows an area dependence for the pumping intensity $I \propto A^{-0.52}$ that agrees with the cases $\eta = 1, 0.3$ and does not too strongly deviate for $\eta = 3$. The lasing threshold is different from sample to sample. For $\eta = 1$ its standard deviation is about half of its absolute value for all examined N. Thus our model predicts the qualitatively valid behavior of the lasing threshold. Unfortunately, the number of particles in experiment always exceeds 5000, so we can directly compare only the extrapolations.

The observed sensitivity of the lasing threshold to the size of the planar sample of fixed density proves the formation of collective optical modes [3,5]. This process reaches maximum efficiency when all local particle resonances occur at the same frequency as in the case we have considered. The similarity between experimental and theoretical lasing threshold behaviors argues that deviations of local resonant frequencies from each other is not very significant. We found that the lasing threshold becomes almost size independent when frequencies deviate from each other by more than the near-neighbor dipolar coupling $\sim nd^2$ [Eq. (2)] (n is the density of particles, for real scattering particles one can express this coupling as the product of the width of the resonance and the density parameter $n\lambda^3$), contrary to the experiment, while the dependences shown in Fig. 1 hold in the opposite limit. Therefore we believe that our choice of equal frequencies $\omega_k = \omega_0$ for all N particles is qualitatively justified.

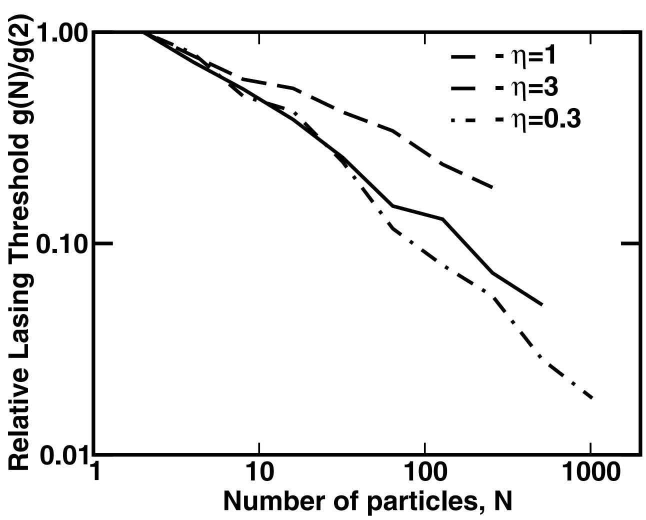

FIG. 1. Dependence of the lasing threshold on the number of particles (sample area).

We begin the discussion of our results phenomenologically. Our approach is qualitatively similar to [14]. One can make the assumption that the eigenmodes of the system (1) and (2), taken at zero gain, can be characterized by the distribution $P(\gamma)$ of their decay rates γ. Then the gain g required to get lasing can be approximately defined as the minimum decay rate taken over all N lossy modes [5,14]. The probability $F(g)$ that the lasing threshold is given by some particular value of the decay rate g (between g and $g + dg$) is composed of the product of two probabilities that one of the N modes has a decay rate within the above interval $NP(g)dg$ and others have bigger decay rates $[\int_g^\infty P(x)\,dx]^{N-1}$. In the limit of large N one can express the function F in the approximate form

$$F(g) \approx NP(g)\exp\left(-N\int_0^g P(x)\,dx\right). \quad (3)$$

If one assumes that, at the fixed density (η), $P(g)$ is a universal scale invariant function $P(g) \propto g^{\alpha}$ weakly dependent on N at small decay rates, the mean lasing threshold and its relative fluctuation can be computed making use of Eq. (3),

$$\langle g\rangle = \frac{1}{N^{1/(\alpha+1)}}(\alpha + 1)^{1/(\alpha+1)}\Gamma\left(\frac{2+\alpha}{1+\alpha}\right),$$
$$\Delta = \frac{\delta g}{g} = \sqrt{\frac{\Gamma[(3+\alpha)/(1+\alpha)]}{\Gamma[(2+\alpha)/(1+\alpha)]^2} - 1}\,. \quad (4)$$

If we take $\alpha = 1$, we obtain the expected behavior $g \propto 1/N^{1/2}$ for the lasing threshold, and relative fluctuation would be $\Delta \approx 0.52$ in agreement with our study. One should note that the fluctuations become smaller when the interparticle distance is reduced. The analysis in detail and the extensive experimental investigation is in progress. The results would be reported elsewhere.

Thus the observed behavior of the lasing threshold can be due to fluctuations. Independence of the distribution $P(g)$ parameters of the particle number N implies that the random cavity size (size of the most weakly decaying quasistate) is smaller than the size of the whole system. We have studied high quality collective modes and have seen that they, in fact, occupy a few scatters (e.g., from 5 to 10 for $N = 100$), and therefore the phenomenological description (4) is justified qualitatively. The lasing threshold decreases with the sample size because of the increase of the probability for optimum configurations of particles (high quality cavities) rather than the formation of larger cavities. Thus the design of an efficient laser does not require a large sample but rather the special arrangement of particles leading to the optimum fluctuation.

The simulations do not show the presence of the gap [14] in the distribution of the loss rate g at low g comparable to average thresholds found in our study (Fig. 1). The gap should lead to saturation in the lasing threshold dependence on N at a disk size around several wavelengths, contrary to our simulations. The strong reduction of the gap size can be due to the specifics of the dipole-dipole interaction (2) having important correlations for different centers that are absent in the random matrix model; cf. [20]. The modes characterized by very long lifetime, filling the upper part of the gap, can be formed from destructive interference of collective dipolar oscillations in the regime of large optical density (small interparticle distance compared to the wavelength). In fact, the increase of the wavelength (decrease of η) at fixed interparticle distance leads to decreasing the threshold (see Fig. 1). The explicit realization of the regime studied in [20], i.e., 3D ensemble of resonant particles at the high density n ($n\lambda^3 \gg 1$) should form more efficient cavities with the decay rate distribution $P(g) \propto g^0$ (see Ref. [20]) leading to the lasing threshold behavior $\langle g \rangle \propto 1/N$ [cf. Eq. (4) with $\alpha = 0$]. Such a system can be formed by metal particles having a plasmon resonance at wavelength much larger than the particle size [16]. The formation of high quality cavities found in [16] possibly can be explained using the technique developed in [20] and these cavities are very promising for efficient lasing. One should note that the reduced dimensionality of the problem can also affect the properties of modes.

As a possible alternative one should mention the results of experimental and theoretical studies of delay time statistics for light transmission in the random quasi-one-dimensional medium [21]. It can be interpreted [5] as the decay rate distribution $P(g) \propto g$ ($g = 1/\tau$). This law is in accord with our results [case $\alpha = 1$, Eq. (4)] and experimental behavior. However, it is not clear how to justify theory arguments [21] for vector electric field of the lossy planar system studied in our case.

We suggest a working model to describe the lasing action in a random medium that agrees qualitatively with the experiment for planar ZnO powder. The strong dependence of the lasing threshold on the system size can be explained by fluctuationally formed small size, high quality cavities. An efficient laser needs the optimum configuration of scattering particles, realizing a random cavity that can have very small size. The lasing efficiency can be increased using scattering particles of very small size compared to the resonant wavelength (possibly metallic particles with a surface plasmon resonance).

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