Statistical Properties of One-Dimensional Random Lasers

Oleg Zaitsev,[1,*] Lev Deych,[2,†] and Vladimir Shuvayev[2]

[1]*Fachbereich Physik, Universität Duisburg-Essen, Lotharstrasse 1, 47048 Duisburg, Germany*
[2]*Physics Department, Queens College of City University of New York, Flushing, New York 11367, USA*
(Received 12 August 2008; published 30 January 2009)

Statistical properties of a laser based on a one-dimensional disordered superlattice open at one side are studied numerically. The passive normal modes of the system are determined using the Feshbach projection technique. It is found that the mode competition due to the spacial hole burning leads to a saturation of the number of lasing modes with increasing pump rate. It is also responsible for non-monotonic dependence of intensities of lasing modes as functions of pumping. Computed distributions of spectral spacing and intensity statistics are in qualitative agreement with experimental results.

Random lasers [1] comprise a large variety of active radiative systems that are based on disordered media. They possess one or more characteristics that distinguish them from the traditional lasers such as absence of the resonator, irregular electric field distributions and spectra, etc. A number of problems can be formulated in connection with random lasers. One group of questions is related to light localization in disordered systems with amplification and absorption [1]. Another class of problems deals with statistics of lasing modes [2–6], e.g., distribution of their frequencies, intensities, etc. From a methodological point of view, extension of standard laser theory to random lasers is an important open question, although some progress in this direction has been made [2,4,7,8].

Statistical properties of random lasers were investigated in several theoretical and experimental papers. Experimentally, distributions of the spacial size of the modes and of the spacing between lasing frequencies in porous GaP were reported in Ref. [9]. The spacing statistics were also studied in lasers based on colloidal solutions [3], where, in addition, statistics of the emitted intensity were measured.

Theoretical studies of this issue were mainly performed for lasers with chaotic resonators, which are similar, in certain aspects, to random lasers. In Ref. [2] random-matrix theory was used to describe weakly open chaotic cavities, where the average number of lasing modes for a given pump strength and the probability distribution of the lasing thresholds were derived. In Refs. [4–6] the average number of modes, its fluctuations, and spectral spacing statistics were computed numerically, particularly, for the case of cavities with overlapping resonances. Statistics of light reflection by a random laser were studied in Ref. [10].

In this Letter we study statistical properties of a particular model of random lasers from first principles, without relying on random-matrix-type hypothesis about statistical properties of cavity modes. Regular semiclassical multi-mode laser theory [11,12] was first applied to lasing in wave-chaotic resonators in Ref. [2]. However, in the case

of lasers based on disordered structures, this theory has to be modified to take into account several factors specific for these systems. First, in this case the openness of the system is essential and should be properly incorporated into the theory. In Ref. [13] the modes of the open cavities were considered using Feshbach projection technique [14], and this approach was applied specifically to random laser systems in Ref. [4]. An alternative approach based on so-called "constant-flux modes" was developed in [8]. Second, the inhomogeneity of the background dielectric constant introduces gain-induced coupling between modes, which is present already in the linear approximation [7], but can be enhanced due to nonlinear hole burning effects [15].

In this work we rederive Maxwell-Bloch lasing equations in the third order of nonlinearity rigorously taking into account the openness of the system. These equations are used for numerical statistical analysis of lasing from a one-dimensional disordered structure. We apply the Feshbach method to compute complex eigenfrequencies and wave functions of the structure under consideration. It is our main conclusion that the correlation of the wave functions critically affects lasing properties of the system. In particular, they are responsible for saturation of the number of lasing modes and a nonmonotonic behavior of lasing intensities with increasing pumping. Both these effects are absent in the chaotic cavities described by random-matrix theory, in which wave functions are not correlated [2,4]. A similar nonmonotonic behavior of intensities was recently found in Ref. [15] using a different approach. We also calculated statistical distributions of spectral spacings and mode intensities, which show qualitative agreement with experiment.

We consider a one-dimensional structure open at one side, which is characterized by a real nonuniform refractive index $n(x)$ for $0 \leq x \leq L$, $n = 1$ for $x > L$, and an ideal mirror at $x = 0$. Normal modes of this system have a finite lifetime even in the absence of absorption. They can be found using the system-and-bath approach [13]. This ap-

proach requires division of the entire system in a closed resonator and environment, which interact through specified boundary conditions. In our formulation we designate the region $0 \le x \le L$ as a resonator with Dirichlet boundary condition at $x = L - 0$, then the bath region $x > L$ must be described by Neumann conditions at $x = L + 0$ [16].

If eigenfrequencies ω_λ^c and eigenfunctions $\psi_\lambda^c(x)$ of the closed resonator are known, then the true normal modes of the open system are obtained by diagonalizing the non-Hermitian matrix

$$\Omega(\omega) = \Omega^c - i\pi W(\omega)W^\dagger(\omega). \tag{1}$$

Here Ω^c is a diagonal matrix of the eigenfrequencies ω_λ^c and $W(\omega)$ is a column of coupling elements of the modes λ with the δ-normalized bath modes $\sqrt{2/\pi}\cos[\omega(x - L)]$ (in the system of units with the speed of light $c = 1$). Explicitly, $W_\lambda(\omega) = (\psi_\lambda^c)'(L)/\sqrt{2\pi\omega_\lambda^c\omega}$. The eigenvalues

of $\Omega(\omega)$, $\Omega_k(\omega) \equiv \omega_k(\omega) - i\kappa_k(\omega)$, provide frequencies and decay rates of the normal modes. Since $\Omega(\omega)$ is non-Hermitian we have to differentiate between left, $|l_k(\omega)\rangle$, and right, $|r_k(\omega)\rangle$, eigenvectors, which are biorthogonal. They can be used to define left and right eigenfunctions $\psi_k^j(x; \omega) \equiv \sum_\lambda \psi_\lambda^c(x)\langle\lambda|j_k(\omega)\rangle$, $j = l, r$, of the open system. The electric field $E(x; \omega)$ in the frequency representation and other relevant functions can be expanded in terms of these normal modes as $E(x; \omega) = \sum_k E_k(\omega)\psi_k^r(x; \omega)$.

The gain medium is described by the polarization and the population difference that interact with the classical field in the resonator. The atomic variables can be eliminated perturbatively from the coupled equations of motion [17]. Following Ref. [18] we carried out this procedure in frequency domain, which is more convenient in our case than more traditional time-domain consideration. The resulting equations for mode amplitudes are

$$-i[\omega - \omega_k(\omega) + i\kappa_k(\omega) - ipD(\omega)]E_k(\omega) = \frac{1}{2}pD(\omega)\int\frac{d\omega'd\omega''}{(2\pi)^2}D^\|(\omega')\sum_{k_1,k_2,k_3}E_{k_1}(\omega - \omega')$$

$$\times[B_{kk_1k_2k_3}(\omega, \omega - \omega', \omega' - \omega'', -\omega'')D^*(-\omega'')E_{k_2}(\omega' - \omega'')E_{k_3}^*(-\omega'')$$

$$+ B_{kk_1k_3k_2}(\omega, \omega - \omega', -\omega'', -\omega' - \omega'')D(-\omega'')E_{k_3}(-\omega'')E_{k_2}^*(-\omega' - \omega'')], \tag{2}$$

where the right-hand side (r.h.s.) contains nonlinear terms in the leading (third) order in the field. In these equations, p is a pump-rate parameter (uniform pumping is assumed), $D(\omega) \equiv [1 - i(\omega - \nu)/\gamma_\perp]^{-1}$, $D^\|(\omega) \equiv (1 - i\omega/\gamma_\|)^{-1}$, ν is the atomic-transition frequency, $\gamma_\perp$ and $\gamma_\|$ are polarization and population relaxation rates, and

$$B_{k_1k_2k_3k_4}(\omega_1, \omega_2, \omega_3, \omega_4) \equiv L\int_0^L dx[\psi_{k_1}^l(x; \omega_1)]^*\psi_{k_2}^r(x; \omega_2)\psi_{k_3}^r(x; \omega_3)[\psi_{k_4}^r(x; \omega_4)]^* \tag{3}$$

denote the overlap integrals. The electric field (in the time representation) is measured in units of $E_0 \equiv \sqrt{\hbar\gamma_\perp\gamma_\|/8\pi d^2\nu}$, where d is the atomic-transition dipole moment. The effect of gain-induced linear mode coupling [7] was found to be small in this system and is neglected in Eq. (2). We do not take into account the effects of linear gain and nonlinearity on the lasing frequencies approximating them with solutions of equations $\omega_k(\omega) = \omega$, which will be denoted as ω_k. In the slow-amplitude approximation, the mode amplitudes $E_k(\omega)$ are assumed to be strongly peaked at ω_k. The mode decay rates, eigenvectors, and wave functions in this case can be taken at the respective lasing frequencies, and the frequency arguments can be omitted. We transform Eq. (2) to the time representation and obtain rate equations for the intensities, $I_k \equiv |E_k(t)|^2$, in the form

$$\dot{I}_k = 2I_k\left(p|D_k|^2 - \kappa_k - p\sum_{k'}C_{kk'}I_{k'}\right), \quad D_k \equiv D(\omega_k). \tag{4}$$

Here terms oscillating at beat frequencies have been ignored. The nonlinear coupling between the modes

$C_{kk'} = |D_{k'}|^2\,\mathrm{Re}[B_{kk'k'k}D_k]$ depends on the overlap integrals (3). This is a simplified expression for $C_{kk'}$, accurate when the population inversion is time independent. The correction due to population pulsations, $\Delta C_{kk'} = \frac{1}{2}\,\mathrm{Re}[B_{kk'kk'}D^\|(\omega_k - \omega_{k'})D_k(D_k + D_{k'}^*)]$ ($k \ne k'$) [6], can be disregarded for typical situation $\gamma_\| \ll \gamma_\perp$. The r.h.s. of Eq. (4) has a transparent physical meaning of a balance between the gain, damping, and nonlinear saturation that prevents the intensity from growing indefinitely.

The amplitude of the emitted modes outside of the resonator, e_k^{out}, is related to the internal mode amplitudes, E_k, by the input-output relation [13] with zero input: $e_k^{\mathrm{out}}(t) = -iE_k(t)W^\dagger|r_k\rangle$. Using this relation in combination with natural representation of matrix (1) as a sum of Hermitian and anti-Hermitian matrices, one can derive a physically transparent expression relating the inside and outside intensities:

$$I_k^{\mathrm{out}} = \kappa_k I_k/\pi. \tag{5}$$

We model a one-dimensional disordered resonator by a superlattice $ABAB\ldots$ consisting of N_l layers of fixed width

a. All layers A have the same refractive index n_A and for layers B the index is drawn randomly from an interval $(n_{\min}, n_{\max})$. In all our numerical examples the parameters are $a = 1$, $N_l = L/a = 200$, $n_A = 1.5$, $n_{\min} = 0.9$, and $n_{\max} = 1.3$ (the refractive index is defined relative to the surrounding medium, where it is set to unity). This system is a periodic-on-average structure with remnants of the bands of a periodic lattice with $N_l/2$ elementary cells. While the band gaps are washed out by disorder we can still define the boundaries of (quasi)bands as minima of the disorder-averaged localization length (see inset of Fig. 2). The number of eigenmodes ω_λ^c for each band fluctuates from realization to realization around $N_l/2$. We focus our simulations on the frequency region of the third band $2.40 \leq \omega_\lambda^c \leq 3.61$, and, respectively, restrict the basis of eigenvectors $\psi_\lambda^c(x)$ used to construct matrix (1) to the modes from this band.

We computed numerically the stationary solutions ($\dot{I}_k = 0$) of Eq. (4) which satisfy the condition $I_k(p) \geq 0$ while continuously increasing pumping from zero. It should be understood that this method does not allow us to detect situations when a mode loses its stability without passing through zero intensity.

The results presented below are obtained with 1700 realizations of disorder. Figure 1 shows the pump dependence of the average number of modes $\langle N \rangle$ and its relative fluctuations $\sqrt{\mathrm{var}(N)}/\langle N \rangle$. The atomic frequency ν is chosen at the middle of the band and the gain width $\gamma_\perp$ is equal to 20% of the bandwidth. An important effect revealed in this figure is the saturation of $\langle N \rangle$ for large p at the level of 44 modes. This phenomenon is not an artifact of the finite basis, which contains 100 available modes. The saturation appears as a result of the nonmonotonic pump dependence of intensities $I_k(p)$ and mode suppression (see the inset). The nonmonotonic pump dependence was observed numerically in two-dimensional disordered
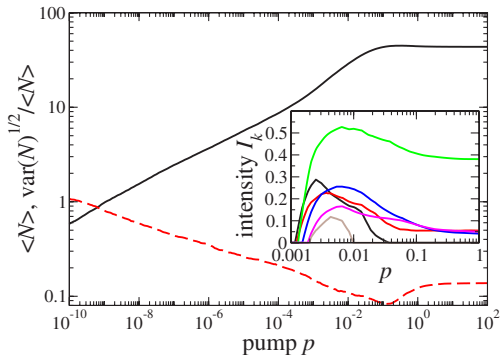
lasers as well [15]. Since in systems with uncorrelated wave functions, such as two-dimensional chaotic resonators, where $B_{kk'k'} = 1 + 2\delta_{kk'}$, mode suppression does not occur [2], we relate the origin of the saturation effect to statistical correlations between modes manifested via the overlap integrals $B_{kkk'k'}$. This effect should not be confused with saturation of the number of lasing modes in the regime of strong localization [19], where saturation is due to "exhaustion" of the number of available nonoverlapping localized modes. In the situation considered here the system is in a truly multimode regime with overlapping modes and the saturation is a result of a nontrivial interplay between effects of self- and cross-saturation.

Before saturation the variance of the number of modes behaves as $\mathrm{var}(N) \propto \langle N \rangle^{0.74}$. Deviation from this behavior marks transition to the regime of strong nonlinear mode competition.

Figure 2 displays probability distributions of spacing $\Delta\omega$ between frequencies of the neighboring lasing modes. If the atomic frequency ν is at the band center and $\gamma_\perp$ is rather small (1% of the bandwidth, in this example), then the distribution has three maxima (dashed line). This behavior results from the existence of well localized modes at the both band edges that despite experiencing a small gain still can lase due to their long lifetimes. As a result we have three well separated groups of lasing modes giving rise to three maxima in the spacing distribution. If the gain is centered at the band edge (dotted line), most of the lasing modes come from that edge, but modes with extremely small κ_k at the opposite edge can be excited as well. This produces a second maximum at the full bandwidth (outside of the plot range).
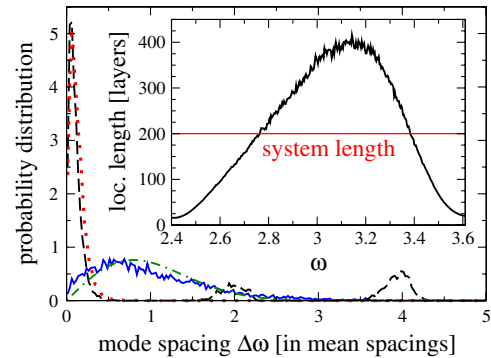

FIG. 1 (color online). Average number of lasing modes $\langle N \rangle$ (solid line) and its fluctuations $\sqrt{\mathrm{var}(N)}/\langle N \rangle$ (dashed line), as a function of the pump rate p. For small p, $\langle N \rangle \propto p^{0.19}$ and $\sqrt{\mathrm{var}(N)}/\langle N \rangle \propto p^{-0.12}$. Inset: Intensities $I_k(p)$ of several lasing modes excited in the intermediate pump range, for one disorder realization. Parameters: $\nu = 3.005$ and $\gamma_\perp = 0.242$.

FIG. 2 (color online). Probability distribution of the spectral spacing between lasing modes ($\gamma_\perp = 0.0121$, $p = 10^{-3}$). Parameters and properties for the dashed line, dotted line, and solid line are, respectively, atomic frequency $\nu = 3.005, 2.405, 2.405$; modes collected from the frequency interval $(2.400, 3.610)$, $(2.400, 3.610)$, $(2.400, 2.521)$; determined mean spacing $\overline{\Delta\omega} = 0.27, 0.18, 0.019$. Dash-dotted line: Wigner surmise (see the text). Inset: Average localization length (in units of the layer width) in the third quasiband, as a function of eigenfrequency ω. Horizontal line indicates current system size.
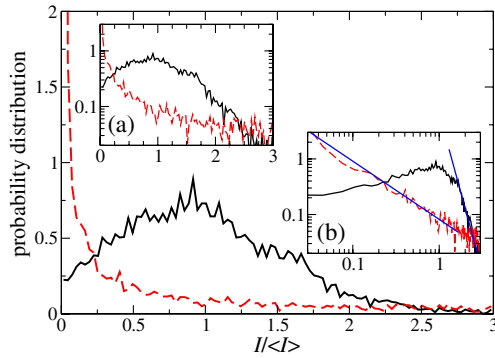
FIG. 3 (color online). Probability distributions $P(I/\langle I \rangle)$ of internal mode intensities I_k (solid line) and output intensities I_k^{out} (dashed line). Parameters: $\nu = 2.405$, $\gamma_\perp = 0.242$, $p = 10^{-3}$; modes are collected from the frequency interval (2.400, 2.521). Mean values: $\langle I_k \rangle = 0.135$, $\langle I_k^{\text{out}} \rangle = 1.78 \times 10^{-6}$. Insets: (a) Semilogarithmic plots, (b) logarithmic plots with the fits $P(x) = 6.6x^{-5.7}$ (internal) and $P(x) = 0.082x^{-1.0}$ (output), where $x \equiv I/\langle I \rangle$.

The solid line in Fig. 2 presents local spacing distribution for modes taken from a narrow spectral strip (10% of the bandwidth) at the band edge, ν being within the strip. This distribution displays mode repulsion (vanishes in the limit $\Delta \omega \to 0$) and is similar to the Wigner surmise $P_W(\Delta \omega) = (\pi \Delta \omega / 2) \exp(-\pi \Delta \omega^2 / 4)$ (dash-dotted line), which approximates the spacing distribution in passive closed chaotic systems. The repulsion occurs because the length L is short enough, so that even states with localization length of $L/10$ can spatially overlap. Moreover, two nonoverlapping modes may have close frequencies, but one of them will be localized closer to the opening and suppressed for moderate pump strength.

Probability distributions for internal and output intensities of lasing modes are shown in Fig. 3. The output distribution has a singularity at zero intensity, whose existence is related to a very broad (several orders of magnitude) distribution of the decay rates of the modes in our system. Indeed, the modes with smaller decay rates κ_k are preferentially excited, but have lower output intensities according to Eq. (5). Both distributions have approximately a power-law asymptotic behavior at large intensities, with different exponents (see the insets).

It is interesting to compare our numerical results for the spectral spacing (Fig. 2) and the intensity distributions (Fig. 3) with available experimental data. The spacing distribution in porous GaP was found to be well approximated by the Wigner surmise [9], while in colloidal solutions of TiO_2 particles the mode repulsion was not of Wigner type [3]. At the same time, the power-law distribution of intensities of lasing modes found in Ref. [3] agrees with our simulations.

In conclusion, we studied numerically a model of disordered laser based on a one-dimensional open resonator. The passive normal modes of the system were determined self-consistently using the Feshbach projection technique. The intensities of lasing modes were found from the rate equations within the semiclassical third-order laser theory. Mode competition, as a consequence of the spacial hole burning, leads to nonmonotonic pump dependence of intensities and mode suppression. The number of lasing modes saturates with increasing pump rate. The local spectral spacing distribution shows a Wigner-like mode repulsion. Globally, the distribution can have several maxima due to the quasiband structure of the spectrum. Distributions of the mode intensities have a power-law asymptotic tail. Output intensities are distributed over several orders of magnitude, reflecting the spread of radiative lifetimes of normal modes.

*oleg.zaitsev@uni-duisburg-essen.de
†lev.deych@qc.cuny.edu

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