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REVIEW ARTICLE

Recent developments in the theory of multimode random lasers

Oleg Zaitsev¹ and Lev Deych²

- ¹ Fachbereich Physik, Universität Duisburg-Essen, Lotharstraße 1, 47048 Duisburg, Germany
- 2 Physics Department, Queens College of City University of New York, Flushing, NY 11367, USA

E-mail: oleg.zaitsev@uni-duisburg-essen.de and lev.deych@qc.cuny.edu

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Abstract

We review recent extensions of semiclassical multimode laser theory to open systems with overlapping resonances and inhomogeneous refractive index. An essential ingredient of the theory is a system of biorthogonal quasimodes that describe field decay in an open passive system and are used as a basis for lasing modes. We discuss applications of the semiclassical theory, as well as other experimental and numerical results related to random lasing with mode competition.

Keywords: random lasers, semiclassical laser theory, quasimodes

1. Introduction

The term 'random lasing' encompasses a number of phenomena related to light amplification in systems characterized by a spatial distribution of the electromagnetic field which is much more complex (irregular) than for well-defined cavity modes of standard lasing structures. Most directly this term describes emission of light by spatially inhomogeneous disordered materials not bounded by any artificial mirrors, even though it is also used in the case of systems with well-defined cavities, which, however, are characterized by chaotic ray dynamics. These two classes of random lasers are significantly different, but in some situations, when the statistical properties of the field in disordered systems and in chaotic cavities are similar, the emission properties of the two classes of lasers can be discussed on an equal footing [1]. In the course of the last decade, random lasing has been observed in many different kinds of disordered materials (polymer films [2], porous materials [3], powders [4], ceramics [5, 6], clusters [7], colloidal solutions of nanoparticles [8]), so it can be regarded as a universal property of disordered structures.

Lasing in any system is produced due to a combination of two factors: optical amplification and feedback. In ordinary lasers for the presence of a feedback one always assumes the existence of well-defined phase relations between

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waves propagating in opposite directions. Put differently, the presence of a feedback is described as the existence of welldefined long-living cavity modes characterized by a regular spatial pattern of the electromagnetic field, which sets up inside the lasing structure in the stationary regime. In [9, 10], where the concept of random lasers was conceived, a possibility of lasing caused by a different type of a feedback was proposed. It was shown that even if propagation of light is described in terms of a diffusion equation, which completely ignores its wave nature and does not have any phase information, one can still have a laser-like behaviour of emission characterized by a threshold, spectral narrowing, relaxation oscillations and other attributes of laser oscillations. The physical origin of this phenomenon lies in a significant increase of the length of the light trajectory inside the finite amplifying volume due to multiple scattering. Since amplification results in exponential growth of light intensity with the distance travelled inside the gain medium, it can be characterized by the gain length $L_{\rm g}$, which depends on the gain factor and the diffusion coefficient of light in the medium. The transition to lasing occurs when the gain length exceeds the loss length, L_1 , so the threshold condition can be written down as $L_g = L_1$. This type of feedback is called incoherent or nonresonant feedback. The latter term refers to the absence of any resonant features in the distribution of the field inside

the gain medium or, in other words, the absence of any distinct modes: the spatial distribution of light intensity is the same regardless of its frequency. In this situation the lasing frequency is determined by only one remaining resonant element of the system—the atomic transition, so the singlepeak emission spectrum is the characteristic feature of lasing with nonresonant feedback [9, 10]. The authors of [11, 12], where nonresonant feedback was discussed in connection with lasing in cavities with rough surfaces, formulated four conditions required for realizing such completely nonresonant situations: (i) no mode degeneracy, (ii) equal mode loss, (iii) mode overlapping (the resonance width of each mode due to radiative losses must be larger than the mean intermode spacing) and (iv) mode mixing (there must be processes causing the frequency of emitted photons to change by an amount larger than the spectral distance between the modes). While conditions (i), (iii) and (iv) are realized automatically in disordered systems of large enough dimensions and chaotic cavities, the condition (ii) is more difficult to fulfil. Indeed, the distribution of widths of scattering resonances in chaotic cavities and disordered systems was found to be rather broad with different modes having decay rates differing by orders of magnitude [13]. However, when interest in random lasers was renewed in 1994 following the observation of lasing from solutions of dye molecules surrounded by titanium dioxide particles [14], the importance of this condition was apparently underappreciated. Namely, results of [14] and subsequent experiments were usually explained on the basis of the concept of nonresonant feedback with its underlying assumption that the transport of light can be described within the diffusion approximation (see, e.g., [15–19]). The validity of the diffusion approximation is usually associated with weak scattering of light, when the system is far away from the Anderson localization transition and interference effects are weak. Therefore, nonresonant or incoherent feedback was by extension associated with the regime of weak scattering of light (see, e.g., [20, 21]).

Inducing stronger light scattering by increasing the concentration of scatterers, it became possible to observe a qualitatively new phenomenon [4, 22–24]: with increasing scattering, multiple emission lines appeared in the spectrum, instead of just a single peak at weaker scattering. New peaks, characterized by much narrower linewidths, emerged one by one with increasing pumping. Since incoherent feedback can only result in a single-frequency emission spectrum, it was suggested in [4] that the changes in the emission spectrum are due to the transition from incoherent to coherent feedback. This idea was supported by studies of photon statistics, which showed that, like for ordinary lasers, light emitted at the peak frequencies had a Poisson photon count distribution [2, 25].

Originally it was suggested in [4] that the feedback is provided by randomly occurring closed trajectories formed by multiply scattered light. Eventually this idea was developed to a more general concept of random cavities (resonators), arising in a strongly scattering medium. Anderson localization was considered as one of the mechanisms that may be responsible for the formation of such cavities [26–28]. In order to verify this assumption a great deal of effort has

been devoted to analysing lasing in one-dimensional models in which all states are localized [29–33]. The localization-based approach allowed us to explain a number of experimentally observed results such as mode repulsion and saturation of the number of lasing modes [34]. However, it has never been convincingly demonstrated that light in strongly scattering three- or two-dimensional samples was indeed close to the Anderson transition. An alternative mechanism of formation of cavities that could be responsible for coherent feedback in random media was put forward in [35], where it was suggested that random fluctuations of the refractive index of a disordered medium can result in macroscopically large ring-like configurations capable of trapping light for long times and serve, therefore, as random resonators. model was supported by studies of lasing in π -conjugated polymers [36-38], where a certain degree of universality in the spectral distribution of lasing modes was found. This universality was explained by noting that among multiple random resonators only those with the largest Q-factors (optimal resonators) gave a major contribution to the lasing. While the distribution of characteristics of the resonators is very broad, the optimal resonators have almost identical characteristics, thus explaining the observed universality. These random light-trapping configurations are analogous to so called prelocalized states, known to exist in the case of electrons in random potentials, and can arise even when a disordered system is far away from the localization transition. It was, however, shown in [35] that the trapping configurations can appear with any appreciable probability only if spatial fluctuations of the refractive index are correlated over large enough distances, which might be a reasonable assumption for the polymer samples studied in [36–38], but is more difficult to justify for the ZnO powders [4]. Yet another alternative model of random lasing was proposed in [39, 40], where random lasers were treated as lasers with distributed feedback. This approach is somewhat similar to the random resonator model one, the only difference being that, instead of dealing with ring-like resonators, the distributed-feedback model assumed that large-scale almost periodic Bragg-like configurations are responsible for the lasing. This model, however, has not yet been sufficiently developed.

More recent developments in the field of random lasers have been associated with renewed attention to the weakly scattering samples. An emission spectrum containing very sharp spikes was observed for very weakly scattering samples in [41]. It was argued in that paper that the appearance of the observed peaks did not require any feedback and could be explained by assuming that some of the spontaneously emitted 'photons' travel much longer distances than the average ones. This idea was supported by simulations based on the random walk model (hence the quotation marks on the 'photon'), which reproduced the experimental data fairly well. However, it was found in [42, 43] that there are two kinds of lines in the emission spectrum of a random laser. One, called the 'spikes' in [42, 43], similar to that observed in [41], was detected (with strong enough pumping) even for samples without any scatterers at all. The second type, called the 'peaks', only appeared in the presence of scatterers. The

spikes and peaks demonstrated significantly different statistical properties, which allowed the authors of [42, 43] to attribute the former to amplified spontaneous emission and claim that only the latter correspond to true lasing with coherent feedback. The significance of these developments consists in the realization of the fact that weak scattering by itself does not guarantee nonresonant feedback and that the diffusion model may not be applicable to active systems even when scattering is weak. This point was reinforced in [27], where it was shown that even in a weakly scattering active medium characterized by strong radiative leakage, as well as spectral and spatial overlap of the modes, lasing from individual modes can still occur. Clearly, this is only possible because even in a weakly scattering system there exists a broad distribution of radiative lifetimes emphasized by the onset of lasing.

Since observation of the multipeak emission spectrum for weakly scattering systems often required enhanced pumping and its concentration within a rather small volume of the sample, effects of the inhomogeneity of pumping on the lasing spectrum have also been studied, in [8, 42]. There it was concluded that the inhomogeneity might play a role in promoting this phenomenon.

As a result of these developments the focus of theoretical research in the field of random lasers has shifted from identifying special configurations responsible for lasing to accepting that a random laser is a multimode system and needs to be treated within the framework of a complete multimode lasing theory. This theory can be developed along the lines of standard semiclassical lasing theory, but it has to incorporate such features specific to random lasers as a much larger role of radiative leakage of the modes and irregular spatial dependence of the refractive index. While this theory is far from being complete, a review of recent developments in this area appears to be useful. Presenting such a summary is the main objective of this work, whose structure is as follows. In section 2 we discuss several alternative ways to introduce a modal description of strongly open systems. After presenting the formal multimode theory in section 3, we turn to discussion of recent experimental and numerical results in section 4.

2. Modes of open systems

The problem of introducing a modal description for open systems has a long history. This problem is important not only for the physics of lasers, but also as a first necessary step toward quantizing the electromagnetic field in open resonators. It is not surprising, therefore, that there have been many alternative attempts to introduce a system of modes suitable for a separation of time and coordinate dependences of various physical quantities such as electric or magnetic fields. The difficulty of the problem stems from the fact that openness makes the problem non-Hermitian. Therefore, the standard recipes for introduction of modes and quantization based on eigenvectors of Hermitian operators are not applicable in this situation. In this section we review a few alternative methods for defining electromagnetic modes of open systems suggested by various authors.

In the Coulomb gauge $\nabla \cdot [\epsilon(\mathbf{r}) \mathbf{E}(\mathbf{r}, t)] = 0$, the electric field $\mathbf{E}(\mathbf{r}, t)$ is governed by a wave equation

$$\epsilon(\mathbf{r}) \frac{\partial^2}{\partial t^2} \mathbf{E} + \nabla \times (\nabla \times \mathbf{E}) = 0, \tag{1}$$

where Gaussian units with the velocity of light in vacuum c=1 are used. Often, for the sake of simplicity, the vector nature of the electromagnetic field is neglected, which is possible if the coupling between various polarizations in an open spatially inhomogeneous system is insignificant. Then the electric field is described by the scalar wave equation

$$\epsilon(\mathbf{r})\frac{\partial^2}{\partial t^2}E - \nabla^2 E = 0. \tag{2}$$

In this section we consider unloaded, or passive, systems (without gain) and assume that all the decay of the field is due to the openness, so that the dielectric constant $\epsilon(\mathbf{r})$ is real. In order to demonstrate the variety of approaches developed for dealing with this problem we will discuss Fox–Li modes in section 2, quasimodes in section 2.2, the system-and-bath approach in section 2.3, and the most recent development, constant-flux modes, will be considered in section 2.4.

2.1. Transverse Fox-Li modes

Random lasers are, of course, not the first type of system in which radiative losses cannot be neglected. Historically earliest, the problem of introducing a modal description in the presence of strong radiative losses arose in connection with lasing properties of so called unstable resonators [44]. Fox and Li [45] suggested defining a mode of such a system as a field distribution which reproduces itself after the wave makes one complete round trip inside the resonator. One assumes that there is a well-defined propagation direction of the wave inside the resonator, so that a solution of (2) can be written in the form

$$E(\mathbf{r}, t) = \text{Re}\{E(\mathbf{r}) \exp[i(kz - \omega t)]\},\tag{3}$$

where $k = \omega \sqrt{\epsilon} \ [\epsilon(\mathbf{r}) = \mathrm{const}]$ and $E(\mathbf{r})$ changes on a scale much longer than k^{-1} . The distribution of the field in the transverse direction, $E(\mathbf{r})$, is characterized by solutions of the integral equation

$$\int K(\mathbf{r}_{\perp}, \mathbf{r}'_{\perp}, z) \, \psi_n(\mathbf{r}'_{\perp}, z) \, d\mathbf{r}'_{\perp} = \lambda_n \psi_n(\mathbf{r}_{\perp}, z), \qquad (4)$$

which formally expresses the idea of the field reproducibility and whose kernel is determined by optical characteristics of the resonator. This equation has the form of an eigenvector equation for a non-Hermitian linear integral operator, whose eigenvalue $|\lambda_n| < 1$ describes radiative losses after a round trip. Since the respective eigenvectors are not orthogonal, one has to introduce an adjoint operator describing propagation in the backward direction as

$$\int K(\mathbf{r}'_{\perp}, \mathbf{r}_{\perp}, z) \, \phi_n(\mathbf{r}'_{\perp}, z) \, d\mathbf{r}'_{\perp} = \lambda_n \phi_n(\mathbf{r}_{\perp}, z). \tag{5}$$

The two families of functions, called Fox-Li modes, are biorthogonal, i.e.,

$$\int \phi_m^*(\mathbf{r}_\perp, z) \, \psi_n(\mathbf{r}_\perp, z) \, d\mathbf{r}_\perp = \delta_{mn}, \tag{6}$$

and can be used to construct a modal expansion of the field

$$E(\mathbf{r}) = \sum_{n} c_n(z) \, \psi_n(\mathbf{r}_{\perp}, z). \tag{7}$$

If the ψ_n are normalized to unity, then

$$K_n \equiv \int |\phi_n(\mathbf{r}_{\perp}, z)|^2 d\mathbf{r}_{\perp} \geqslant 1.$$
 (8)

It was shown [44, 46] that K_n is the Petermann excessnoise factor [47], which describes the effect of the openness of the resonator on the fundamental Schawlow–Townes linewidth. While the Fox–Li modes played an important role in understanding unstable resonators, their application to random lasers and especially to the problem of field quantization is rather limited.

2.2. Quasimodes

The idea of quasimodes was transferred to optics from quantum physics. In order to describe scattering resonances in atomic and molecular physics, it was proposed to solve the Schrödinger equation with boundary conditions at infinity containing only outgoing waves and no incoming incident waves (so called Siegert or Gamow boundary conditions) [48]. The solutions of the resulting non-Hermitian eigenvector problem are characterized by complex eigenvalues and eigenvectors diverging at infinity. The latter circumstance makes it impossible to use these modes as a basis for representation of the lasing field or to develop a quantization procedure representing the field in the entire space. At the same time, it was argued in [49] that under certain realistic conditions it is possible to use these modes to represent the field inside the resonator. For both lasing and quantization problems the field is needed everywhere; therefore there have been several attempts to reformulate this problem in a way which would produce a meaningful system of basis vectors valid over the whole space.

One of the approaches, developed in [50], uses modes obeying Siegert–Gamow boundary conditions to present the field inside the cavity, but uses a different set of functions to describe the outside field in order to avoid the divergency problem. These authors argue that such modes describe 'natural' evolution of the field inside the cavity after it was created and allowed to evolve freely and, therefore, call them natural modes. The authors employ an original definition of the biorthogonal inner product by presenting left- and right-propagating components of these modes as components of a two-dimensional spinor. This construction was studied in more detail for a one-dimensional cavity of constant refractive index open at one side and having a perfect mirror at the other side. Its eigenfunctions satisfying the outgoing-wave boundary conditions are of the form $\psi_n(x) \propto e^{i\kappa_n x} + re^{-i\kappa_n x}$, where κ_n

is complex. Its adjoint function $\phi_n(x) = [\psi_n(x)]^*$ matches an incoming wave. The corresponding natural modes are then

$$\Psi_n(x) \propto \begin{pmatrix} e^{i\kappa_n x} \\ re^{-i\kappa_n x} \end{pmatrix}, \qquad \Phi_n(x) \propto \begin{pmatrix} re^{i\kappa_n^* x} \\ e^{-i\kappa_n^* x} \end{pmatrix}.$$
 (9)

Natural modes for the external region are defined to have a real wavenumber k, but, if continued inside the cavity, they would not satisfy the Dirichlet condition at the mirror. Both internal and external modes are biorthogonal to their adjoints in the respective regions of space.

Since the natural modes form complete sets, they can be used to quantize the field over the whole space. Namely, the amplitudes of the modes $\Psi_n(x)$ and $\Phi_n(x)$ become cavity operators a_n and b_n , respectively. Similarly, external operators a(k) and b(k) are defined. The system Hamiltonian

$$H = H_{in}(\{a_n, b_n\}) + H_{out}(\{a(k), b(k)\})$$
 (10)

is a sum of internal and external contributions without crossterms. Coupling between internal and external waves in this formalism arises due to noncommutativity of the internal and external operators. This circumstance makes application of these modes not very convenient; therefore, it would be interesting to try to introduce new commuting internal and external operators. This would result in a Hamiltonian where coupling would enter explicitly in the standard form of crossterms.

2.3. Feshbach projection in the system-and-bath quantization scheme

Field quantization based on quasimodes [50] (section 2.2) is an example of the system-and-bath approach: one introduces separate eigenmodes and field operators inside the open region designated as the 'system' and in the surrounding free space ('bath'). This procedure can be contrasted with the modes-of-the-universe approach, in which the modes are defined over the whole space. The separation into a system and a bath often provides a clearer physical description. For example, the complex energy of a resonator state immediately yields its lifetime, while extracting this information from the real continuous spectrum is less straightforward.

The Feshbach projection technique [51] offers a convenient way to perform the system-and-bath quantization in a rather general setting of equation (1) [52], the only assumption being that $\epsilon(\mathbf{r}) = 1$ outside of a finite domain $\mathcal Q$ of arbitrary shape. The idea of the method is to project the Hilbert space of the modes of the universe into the Hilbert spaces of $\mathcal Q$ and its exterior $\mathcal P$ with appropriate boundary conditions.

Separating variables in (1) with an ansatz $\mathbf{E}(\mathbf{r},t)=\mathrm{Re}[\mathbf{E}_{\omega}(\mathbf{r})\exp(-\mathrm{i}\omega t)]$ and introducing a new, in general, vector-valued function $\boldsymbol{\phi}_{\omega}(\mathbf{r})=\sqrt{\epsilon(\mathbf{r})}\,\mathbf{E}_{\omega}(\mathbf{r})$, we can formulate an eigenvalue problem

$$L\phi_{\omega} \equiv \frac{1}{\sqrt{\epsilon(\mathbf{r})}} \nabla \times \left[\nabla \times \frac{\phi_{\omega}}{\sqrt{\epsilon(\mathbf{r})}} \right] = \omega^{2} \phi_{\omega}$$
 (11)

for the Hermitian operator L. After the projection into the subspaces Q and P, an equivalent system of equations

$$\begin{pmatrix} L_{QQ} & L_{QP} \\ L_{PQ} & L_{PP} \end{pmatrix} \begin{pmatrix} \boldsymbol{\mu}_{\omega} \\ \boldsymbol{\nu}_{\omega} \end{pmatrix} = \omega^{2} \begin{pmatrix} \boldsymbol{\mu}_{\omega} \\ \boldsymbol{\nu}_{\omega} \end{pmatrix}$$
(12)

for the restrictions $\mu_{\omega} = \phi_{\omega}|_{\mathcal{Q}}$ and $\mathbf{v}_{\omega} = \phi_{\omega}|_{\mathcal{P}}$ is obtained. The differential operators $L_{\mathcal{Q}\mathcal{Q}}$ and $L_{\mathcal{P}\mathcal{P}}$ act in their domains and have the same bulk terms as L (11) and interface terms; $L_{\mathcal{Q}\mathcal{P}}$ and $L_{\mathcal{P}\mathcal{Q}}$ act at the interface between \mathcal{Q} and \mathcal{P} . There is a certain freedom in the distribution of the interface terms between the operators, as well as in the division of the whole space between \mathcal{Q} and \mathcal{P} . It is expected, however, that physically relevant quantities will be independent of this choice [53]. The eigenmodes of $L_{\mathcal{Q}\mathcal{Q}}$ and $L_{\mathcal{P}\mathcal{P}}$ are determined from the equations

$$L_{\mathcal{Q}\mathcal{Q}} \boldsymbol{\mu}_{\lambda} = \omega_{\lambda}^{2} \boldsymbol{\mu}_{\lambda}, \qquad L_{\mathcal{P}\mathcal{P}} \boldsymbol{\nu}_{m}(\omega) = \omega^{2} \boldsymbol{\nu}_{m}(\omega), \qquad (13)$$

where ω_{λ} is real and discrete, ω is real and continuous and m is a discrete channel index. The modes are required to obey the boundary conditions that make the interface terms of the operators vanish. For example, in one dimension these could be the Dirichlet/Neumann conditions on the $\mathcal{Q/P}$ side of the interface or vice versa. The eigenfunctions $\mu_{\lambda}(\mathbf{r})$ and $\nu_{m}(\omega; \mathbf{r})$ of the Hermitian operators form complete sets in their domains. Because of the special boundary conditions, an expansion of a mode-of-the-universe ϕ_{ω} in terms of these eigenmodes should deviate strongly from the exact result in a layer around the interface. The width of the layer should go to zero as the number of terms in the expansions becomes infinite.

The field is quantized in each domain separately by assigning the annihilation operators a_{λ} and $b_m(\omega)$ to the eigenmodes μ_{λ} and $\nu_m(\omega)$. The system-and-bath Hamiltonian

$$H = \sum_{\lambda} \hbar \omega_{\lambda} \, a_{\lambda}^{\dagger} a_{\lambda} + \sum_{m} \int \, d\omega \, \hbar \omega \, b_{m}^{\dagger}(\omega) \, b_{m}(\omega)$$
$$+ \hbar \sum_{\lambda m} \int \, d\omega [W_{\lambda m}(\omega) \, a_{\lambda}^{\dagger} b_{m}(\omega)$$
$$+ V_{\lambda m}(\omega) \, a_{\lambda} b_{m}(\omega) + \text{h.c.}], \tag{14}$$

consists of the internal and external contributions and the interaction part with $W_{\lambda m}(\omega) = \langle \boldsymbol{\mu}_{\lambda} | L_{\mathcal{QP}} | \boldsymbol{v}_m(\omega) \rangle$ and $V_{\lambda m}(\omega) = \langle \boldsymbol{\mu}_{\lambda}^* | L_{\mathcal{QP}} | \boldsymbol{v}_m(\omega) \rangle$. In contrast to the case for the Hamiltonian (10), here the system and the bath operators commute and the interaction terms are explicit. From the Heisenberg equations of motion for $a_{\lambda}(t)$ and $b_m(\omega;t)$ one obtains the quantum Langevin equation for $a_{\lambda}(\omega)$ in the frequency representation:

$$i\sum_{\lambda'}[\omega\delta_{\lambda\lambda'} - \Omega_{\lambda\lambda'}(\omega)]a_{\lambda'}(\omega) + F_{\lambda}(\omega) = 0, \qquad (15)$$

where $F_{\lambda}(\omega)$ is the bath-noise operator. The non-Hermitian frequency matrix

$$\Omega_{\lambda\lambda'}(\omega) = \omega_{\lambda}\delta_{\lambda\lambda'} - i\pi(WW^{\dagger})_{\lambda\lambda'}(\omega) - \Delta_{\lambda\lambda'}(\omega), \tag{16}$$

$$(WW^{\dagger})_{\lambda\lambda'}(\omega) \equiv \sum_{m} W_{\lambda m}(\omega) W_{\lambda' m}^{*}(\omega), \qquad (17)$$

contains the imaginary damping term and the real frequency shift, often disregarded. In the rotating-wave approximation, which is applicable when the damping is much smaller than the typical frequencies, the $V_{\lambda m}(\omega)$ contribution can be neglected.

Semiclassically, the electric field in the system (in Q) is $\mathbf{E}_{\omega}(\mathbf{r}) = \sum_{\lambda} a_{\lambda}(\omega) \boldsymbol{\mu}_{\lambda}(\mathbf{r}) / \sqrt{\epsilon(\mathbf{r})}$. Representing the

matrix (16) via its eigenvalues and biorthogonal left and right eigenvectors as $\Omega(\omega) = \sum_{k} |r_k(\omega)\rangle \Omega_k(\omega) \langle l_k(\omega)|$ we can construct quasimodes

$$\psi_{k}(\omega; \mathbf{r}) = \langle \mu(\mathbf{r}) | r_{k}(\omega) \rangle,$$

$$\mathbf{E}_{\omega}(\mathbf{r}) = \sum_{k} \mathbf{E}_{k}(\omega; \mathbf{r}) = \sum_{k} a_{k}(\omega) \psi_{k}(\omega; \mathbf{r}) / \sqrt{\epsilon(\mathbf{r})}$$
(18)

where $a_k(\omega) \equiv \langle l_k(\omega) | a(\omega) \rangle$ and $|\mu(\mathbf{r})\rangle$ [$|a(\omega)\rangle$] is a column of $\mu_{\lambda}(\mathbf{r})$ [$a_{\lambda}(\omega)$]. These modes depend on the real frequency ω imposed by the exterior. With the help of (15) and the bath correlation function at zero temperature $\langle F_{\lambda}(\omega) F_{\lambda'}^{\dagger}(\omega') \rangle = (WW^{\dagger})_{\lambda\lambda}(\omega) \delta(\omega - \omega')$ [54] one obtains the (bath averaged) field correlation in the mode k [55]:

$$\langle \mathbf{E}_{k}(\omega; \mathbf{r}) \cdot \mathbf{E}_{k}^{*}(\omega'; \mathbf{r}) \rangle$$

$$= \frac{\langle l_{k}(\omega) | WW^{\dagger}(\omega) | l_{k}(\omega) \rangle}{[\omega - \omega_{k}(\omega)]^{2} + \kappa_{k}^{2}(\omega)} \frac{| \psi_{k}(\omega; \mathbf{r}) |^{2}}{\epsilon(\mathbf{r})} \delta(\omega - \omega'), \tag{19}$$

where $\omega_k(\omega) \equiv \operatorname{Re} \Omega_k(\omega)$ and $\kappa_k(\omega) \equiv \operatorname{Im} \Omega_k(\omega)$. Thus, the modes $\mathbf{E}_k(\omega; \mathbf{r})$ do indeed describe a leaking field with the frequency given by the equation $\omega = \omega_k(\omega)$ and the decay rate $\kappa_k(\omega)$, as is expected from quasimodes.

2.4. Constant-flux states

So called constant-flux (CF) states were introduced in [56] as an attempt to construct a system of basis functions suitable for modal expansion of the field inside an open resonator, which would also allow us to easily calculate the field outside of the resonator. While these modes are, indeed, very convenient for semiclassical lasing theory, it is not clear at the present time whether they can be used for quantization of the field. The CF modes are very similar to the modes [52] of section 2.3: in the interior of the cavity region $\mathcal Q$ they satisfy the eigenvalue equation

$$\frac{1}{\sqrt{\epsilon(\mathbf{r})}} \nabla \times \left[\nabla \times \frac{\widetilde{\boldsymbol{\psi}}_{k}(\omega)}{\sqrt{\epsilon(\mathbf{r})}} \right] = \widetilde{\Omega}_{k}^{2}(\omega) \, \widetilde{\boldsymbol{\psi}}_{k}(\omega), \qquad (20)$$

where $\widetilde{\Omega}_k$ is an eigenfrequency and in the exterior domain \mathcal{P} the equation for these modes takes the form

$$\nabla \times [\nabla \times \widetilde{\boldsymbol{\psi}}_k(\omega)] = \omega^2 \widetilde{\boldsymbol{\psi}}_k(\omega) \tag{21}$$

where ω is a real external spectral parameter, which does not coincide with the eigenfrequency $\widetilde{\Omega}_k$. However, equations (20) and (21) are complemented by outgoing-wave boundary conditions at infinity and continuity conditions at the boundary of the resonator. One can think of ω as a frequency of some external force exciting the modes of the cavity, or alternatively, as a spectral parameter of a temporal Fourier transform used to convert the problem from the time to the frequency domain. This spectral parameter becomes an integration variable when inverse transformation back to the time domain is carried out. Description of the outside modes using equation (21) not only allows one to introduce a nondiverging energy flux, but also ensures the biorthogonality of the internal modes. Indeed, electromagnetic boundary conditions are of mixed type and contain a dependence on the eigenfrequencies. As a

result the orthogonality between adjoint modes is destroyed, and transition to (21) is a way to restore it. A similar approach was described in the case of elastic waves in [57]. It was shown in [56] that modes defined this way form a complete set in the domain Q, which can be complemented by a biorthogonal set of adjoint modes. The advantage of these states compared to quasimodes obeying Siegert–Gamow boundary conditions is that the former remain finite at infinity and describe a constant flux of energy coming out from the resonator (hence, 'constant-flux' states). There is no need to introduce special outside modes, the exterior field is calculated by simply matching the outgoing field with the cavity field. This is particularly convenient for calculation of light emission from open resonators.

The CF wavefunctions and their biorthogonal adjoint functions $\widetilde{\phi}_k(\omega; \mathbf{r})$ provide a spectral representation of the interior Green function operator [56] satisfying outgoing boundary conditions at infinity, which can be written in the form

$$[G_{QQ}(\omega; \mathbf{r}, \mathbf{r}')]_{\alpha\alpha'} = \sum_{k} \frac{[\widetilde{\boldsymbol{\psi}}_{k}(\omega; \mathbf{r})]_{\alpha} [\widetilde{\boldsymbol{\phi}}_{k}^{*}(\omega; \mathbf{r}')]_{\alpha'}}{\omega^{2} - \widetilde{\Omega}_{k}^{2}(\omega)}.$$
 (22)

The indices α , $\alpha' = x$, y, z label the polarization of the vector-valued field. The Green function satisfying the same boundary conditions can be defined in the system-and-bath approach as [52] $G_{QQ}(\omega) = [\omega^2 - L_{\rm eff}(\omega)]^{-1}$, where the differential operator $L_{\rm eff}$ has the form

$$L_{\text{eff}}(\omega) = L_{QQ} + L_{QP}(\omega^2 - L_{PP} + i\varepsilon)^{-1} L_{PQ}.$$
 (23)

Thus, the $\widetilde{\psi}_k(\omega)$ are eigenfunctions of $L_{\rm eff}(\omega)$ with the eigenvalues $\widetilde{\Omega}_k^2(\omega)$. In fact, it was shown in [53] for the one-dimensional case that the interface terms of $L_{\rm eff}(\omega)$ disappear precisely when the function that it acts upon satisfies the outgoing boundary conditions. The poles of the Green function, which correspond to scattering resonances, are found by analytical continuation of ω into the complex plane such that the equation $\omega^2 = \widetilde{\Omega}_k^2(\omega)$ is satisfied. At the same time, in the case of CF states, ω is always real and, if it is fixed by some external conditions (for instance, it can be the frequency of incident radiation tuned to be in resonance with the cavity mode), the respective resonant eigenfrequency of the CF state obeys a different equation: $\text{Re}[\Omega_k(\omega)] = \omega$.

It is interesting to compare CF modes with those obtained in the system-and-bath approach described in section 2.3. The first step is to write L_{eff} as a matrix in the eigenbasis of L_{QQ} (see (13)):

$$L_{\text{eff}}(\omega) \simeq \Omega_0^2 - 2\sqrt{\Omega_0} [i\pi W W^{\dagger}(\omega) + \Delta(\omega)] \sqrt{\Omega_0},$$
 (24)

where Ω_0 is the real diagonal matrix of ω_{λ} . The difference between the matrix $\Omega^2(\omega)$ (16) and this matrix reads

$$\Omega^{2}(\omega) - L_{\text{eff}}(\omega) = [\Delta\Omega(\omega), \Omega_{0}] + \Delta\Omega^{2}(\omega),$$

$$\Delta\Omega(\omega) \equiv \Omega(\omega) - \Omega_{0},$$
(25)

where the diagonality of Ω_0 was used. This difference is of second order in the small parameters $|\Delta\Omega_{\lambda\lambda'}/|\omega_{\lambda}|$ and $|\omega_{\lambda}-\omega_{\lambda'}|/|\omega_{\lambda}|$. Therefore, the CF eigenfrequencies and eigenfunctions are expected to become close to $\Omega_k(\omega)$ and $\psi_k(\omega)$ as the above ratios decrease.

2.5. Open resonators in random matrix theory

Explicit construction of quasimodes along the lines of sections 2.2–2.4 in irregular systems may require a fair amount of computation. At the same time, one is often interested in a statistical description of ensembles of such systems. It is well known that certain statistical characteristics of chaotic cavities and diffusive media in the short-wavelength limit are universal. This means that these properties depend only on the symmetries and, possibly, the boundary conditions, but not on the details of the spatial distribution of the refractive index. The universality justifies modelling eigenfrequencies and eigenfunctions of these systems using appropriate statistical ensembles of random matrices, which can be easier to handle analytically or numerically than physical systems. A review of random matrix theory (RMT) in open cavities is given in [58].

A convenient starting point is the following, rather general representation of the scattering matrix of an open system:

$$S(\omega) = \mathbb{I} - 2i\pi W^{\dagger} (\omega - \Omega_{\text{eff}})^{-1} W, \qquad \Omega_{\text{eff}} = \Omega_0 - i\pi W W^{\dagger}.$$
(26)

 $S(\omega)$ is an $M\times M$ matrix in the channel space. It transforms a column of incoming amplitudes into a column of outgoing amplitudes in some basis of the channel states. $\Omega_{\rm eff}$ is called the effective Hamiltonian in electronic systems. It is analogous to the frequency matrix (16). Ω_0 is an $N\times N$ Hermitian frequency matrix of the closed resonator; its eigenvalues are the eigenfrequencies of the cavity. The $N\times M$ matrix W describes coupling between the resonator and the open channels. In the limit usually considered in RMT, $N\to\infty$, the frequency dependence of W is neglected.

RMT prescribes that, in order to model a generic chaotic system with time-reversal symmetry, the real symmetric matrix Ω_0 should be taken from the Gaussian orthogonal ensemble. Without loss of generality, the diagonal (off-diagonal) elements of Ω_0 are drawn from a normal distribution with zero mean and the variance of 2/N (1/N). In the limit $N \to \infty$ the eigenvalues are distributed according to the Wigner semicircle law

$$g(\omega) = \frac{1}{\pi} \sqrt{1 - \frac{\omega^2}{4}}, \qquad -2 \leqslant \omega \leqslant 2, \qquad (27)$$

where $g(\omega)$ is normalized to unity. In the absence of direct coupling between the channels, matrix $S(\omega)$ should become diagonal after the ensemble averaging. This can be achieved simply by taking fixed (for all members of the ensemble) elements $W_{nn} \equiv \sqrt{\gamma_n/\pi} > 0$ for $n \leqslant M$ and $W_{nm} = 0$ otherwise (the limit $M \ll N$ is assumed). Thus, the interaction matrix $\pi W W^{\dagger}$ is diagonal with M nonzero elements γ_n . The strength of coupling between the resonator and the continuum is characterized by the transmission coefficients

$$T_n = 1 - |\langle S_{nn}(\omega) \rangle|^2 = 2 \left[1 + \frac{\gamma_n + \gamma_n^{-1}}{2\pi g(\omega)} \right]^{-1}.$$
 (28)

In particular, the coupling is the strongest for $\gamma_n = 1$. Since the coupling depends on the density $g(\omega)$, one has to be careful not to mix the statistics from different regions of ω or introduce appropriate scaling in order to obtain universal results.

Quasimodes of an open chaotic resonator can be modelled via the resonances of $S(\omega)$, which are the eigenmodes of $\Omega_{\rm eff}$. Statistical properties of the resonant wavefunctions will follow from the surmise that values of an eigenfunction of a closed chaotic cavity are uncorrelated Gaussian random variables at points spaced more than a wavelength apart [59].

Several papers extend the calculations [44, 46] of the Petermann factor [47] to chaotic systems, where, in particular, there is no separation into longitudinal and transverse modes. In the case of chaotic cavity with an opening [60, 61], the RMT effective Hamiltonian Ω_{eff} (26) is modified to account for the amplifying medium in the resonator by adding a correction $i\gamma_a/2$ to its eigenvalues, where $\gamma_a>0$ is the amplification rate. As γ_a is increased from zero, the eigenvalues shift upwards in the complex plane (the gain is assumed to be the same for all relevant frequencies). The first eigenvalue touching the real axis defines the lasing threshold. The Petermann factor is shown to be

$$K = \langle l|l\rangle\langle r|r\rangle \tag{29}$$

where $|l\rangle$ ($|r\rangle$) is the left (right) eigenvector of $\Omega_{\rm eff}$ (for $\gamma_{\rm a}=0$). A supersymmetric calculation [61] reveals that $\langle K\rangle$ scales as the square root of the number of channels in the opening. If the system has a time-reversal symmetry, the matrix $\Omega_{\rm eff}$ is symmetric, i.e., W is real. Then one can impose the condition $|l\rangle=|r\rangle^*$, which leads to an alternative expression

$$K_{\text{TRS}} = \langle r | r \rangle^2.$$
 (30)

In a chaotic dielectric resonator (a domain of uniform refractive index n>1 surrounded by a medium with n=1) light can leak anywhere along the boundary. Hence, the number of output channels M scales with the size N of $\Omega_{\rm eff}$, breaking the standard RMT assumption $M\ll N$. A combination of RMT with the Fresnel laws [62] produces a scattering matrix

$$S(\omega) = -R + TF(\omega)[\mathbb{I} - RF(\omega)]^{-1}T, \tag{31}$$

where $F(\omega)$ is the intracavity propagator and R and T are diagonal matrices of reflection and transmission. The Petermann factor is of the form (29), but the eigenvectors refer to the matrix $RF(\omega)$. After a symmetrization procedure, the form (30) can also be obtained if time-reversal symmetry is present. Comparison of the RMT calculations with the quantum-kicked-rotator model indicates loss of universality as n becomes close to unity, i.e., in a strongly open resonator.

It should be noted that the near-threshold treatment [60–62] yields a value of the Petermann factor which is twice as large as the correct result. This happens because phase diffusion (which mainly contributes to the linewidth) is altered at the threshold by the amplitude fluctuations. The correct prefactor was inserted 'by hand' in equations (29) and (30). A linearization of the quantum Langevin equations far above the threshold [63] yields again the result (29), this time with the proper prefactor.

3. Multimode laser theories for open and irregular systems

As was mentioned in section 1, the current trend in the theoretical description of random lasing consists in extending standard semiclassical multimode lasing theory [64, 65] to situations specific to random lasing: modes with a broad distribution of radiative lifetimes and irregular spatial patterns, inhomogeneity of the background refractive index, a large number of lasing modes emitting in the regime when pumping significantly exceeds the threshold value. While it is true that the multimode lasing was considered one of the signatures of coherent feedback, the discussion of this issue in earlier works related to strongly scattering systems was mostly concerned with a rather trivial situation, when multiple modes originated from different non-overlapping cavities (localized states), which were assumed to be in the single-mode regime due to mode competition [30, 32, 34]. The only nontrivial multimode effect reported in strongly scattering systems was observation of mode coupling in [32]; however, an attempt at its explanation using a two-mode lasing theory given in [66] relied on an incorrect form of the rate equations (this point will be explained later in this section).

One of the first attempts to apply multimode lasing theory to chaotic lasers was undertaken in [1], where equations of standard semiclassical laser theory were combined with ideas of random matrix theory concerning statistical properties of eigenmodes of chaotic resonators. Cavities studied in [1] were characterized by relatively long radiative lifetimes of the modes. Statistical properties of random lasers with strong radiative losses were studied in [67–69] using a combination of random matrix theory with the method of Feshbach projection. A statistical study of one-dimensional strongly open random lasers based on calculations of lasing modes 'from first principles' rather than on phenomenological random matrix-type considerations was carried out in [70].

The role of nonuniformity of the refractive index in the formation of lasing modes and its consequences for lasing dynamics was discussed in [71, 72], where the idea that lasing modes can be significantly different from modes of cold cavities and must be determined self-consistently was formulated. Application of the theory [71, 72] to random lasers was, however, limited because of the restriction of the self-consistency requirements to the linear regime only and neglect of radiative losses of the modes. Both these limitations were removed in [56, 73, 74] (see also the recent review article [75]), where the spatial structure of lasing modes and the lasing frequencies were determined self-consistently from fully nonlinear theory. The approach developed in these papers is based on three main ingredients: (i) use of CF states to incorporate radiative losses of the system, (ii) neglecting population pulsation, which allowed one to take into account nonlinear interactions up to infinite order in the field intensity, and (iii) determining of lasing modes and their frequencies, self-consistently. Results of those works revealed the presence of strong effects related to nonlinear interaction between modes in systems with strongly overlapping (both spectrally and spatially) modes. However, since this method is based on

numerical computation of fixed points of a certain nonlinear map, which, as often happens in nonlinear systems, might have multiple stable and metastable points, the usefulness of this approach might be limited to systems with relatively large intermode spacing. Its applicability to truly diffusive random lasers is difficult to assess from the data published in [56, 73–75], since they do not contain any information about the mean free path, $l_{\rm mfp}$, of light in the structures studied. The diffusive regime arises only when the relations $R\gg l_{\rm mfp}\gg\lambda$, where λ is the wavelength of light and R is the size of the sample, are satisfied, which requires much larger samples than the condition $R\gg\lambda$, fulfilled for samples studied in [56, 73–75].

In sections 3 and 4 we derive a semiclassical multimode lasing theory taking into account recent achievements discussed above and present some examples of its application to situations relevant for random lasers.

3.1. Semiclassical laser equations

A starting point for the semiclassical description of lasers [64, 65] is the wave equation for the electric field (1) with the polarization $\mathbf{P}(\mathbf{r}, t)$ as a source that generates the field (c = 1):

$$\epsilon(\mathbf{r}) \frac{\partial^2}{\partial t^2} \mathbf{E} + \nabla \times (\nabla \times \mathbf{E}) = -4\pi \frac{\partial^2}{\partial t^2} \mathbf{P}(\mathbf{r}, t). \tag{32}$$

In the simplest model, the polarization is produced by twolevel active atoms and obeys the equation

$$\left(\frac{\partial^2}{\partial t^2} + 2\gamma_{\perp}\frac{\partial}{\partial t} + \nu^2\right)\mathbf{P} = -2\nu\frac{d^2}{\hbar}\mathbf{E}(\mathbf{r}, t)\,\Delta n(\mathbf{r}, t),\quad(33)$$

where $\Delta n(\mathbf{r}, t)$ is the population-inversion density, d is the magnitude of the atomic dipole matrix element, ν is the atomic transition frequency (homogeneous broadening is assumed) and γ_{\perp} is the polarization decay rate. The population inversion, in turn, depends on the electric field and the polarization,

$$\frac{\partial}{\partial t} \Delta n - \gamma_{\parallel} [\Delta n_0(\mathbf{r}, t) - \Delta n] = \frac{2}{\hbar \nu} \mathbf{E}(\mathbf{r}, t) \cdot \frac{\partial}{\partial t} \mathbf{P}(\mathbf{r}, t).$$
(34)

If the right-hand side vanishes, Δn relaxes with the rate γ_{\parallel} to the unsaturated population inversion $\Delta n_0(\mathbf{r}, t)$, which is determined by the pump.

The coupled equations (32)–(34) yield, in principle, the distribution of the electric field in the system, if $\Delta n_0(\mathbf{r}, t)$ is given. Rather than carrying out the traditional derivation of lasing equations, which is usually done in the time domain, we find it more convenient to proceed using the frequency representation. We introduce the Fourier transforms

$$\mathbf{E}(\mathbf{r},t) = \frac{1}{\pi} \operatorname{Re} \int_0^\infty d\omega \, \mathbf{E}_{\omega}(\mathbf{r}) \, \mathrm{e}^{-\mathrm{i}\omega t}, \tag{35}$$

$$\mathbf{P}(\mathbf{r},t) = \frac{1}{\pi} \text{Re} \int_0^\infty d\omega \, \mathbf{P}_{\omega}(\mathbf{r}) \, e^{-i\omega t}, \qquad (36)$$

$$\Delta n(\mathbf{r}, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \, \Delta n_{\omega}(\mathbf{r}) \, e^{-i\omega t}$$
 (37)

in the form chosen to facilitate application of the rotating-wave approximation. In addition, we assume that the time dependence of the field and polarization is determined by fast oscillations with frequencies which are close to the atomic frequency ν and residual slow time dependence. In the frequency domain this means that only Fourier components \mathbf{E}_{ω} and \mathbf{P}_{ω} with ω in the close vicinity of ν contribute significantly to the dynamics. Therefore, after performing Fourier transformation of (32)–(34), one can neglect terms of the order of $(\omega - \nu)^2$ in $\omega^2 = (\omega - \nu + \nu)^2 \approx \nu^2 + 2\nu(\omega - \nu)$ and write down the resulting equations as

$$-\epsilon(\mathbf{r}) \left(-\nu^2 + 2\nu\omega\right) \mathbf{E}_{\omega} + \nabla \times (\nabla \times \mathbf{E}_{\omega}) = 4\pi \nu^2 \mathbf{P}_{\omega}, \quad (38)$$

$$[-\mathrm{i}(\omega - \nu) + \gamma_{\perp}]\mathbf{P}_{\omega} = -\mathrm{i}\frac{d^2}{2\pi\hbar} \int_0^{\infty} \mathrm{d}\omega' \,\mathbf{E}_{\omega'} \Delta n_{\omega - \omega'}, \quad (39)$$

 $(-\mathrm{i}\omega + \gamma_{\parallel})\Delta n_{\omega} = 2\pi \gamma_{\parallel} \Delta n_0(\mathbf{r}) \,\delta(\omega)$

$$-\frac{\mathrm{i}}{\pi\hbar} \int_0^\infty \mathrm{d}\omega' (\mathbf{E}_{\omega'-\omega}^* \cdot \mathbf{P}_{\omega'} - \mathbf{E}_{\omega'+\omega} \cdot \mathbf{P}_{\omega'}^*), \tag{40}$$

where we assumed that the pump $\Delta n_0(\mathbf{r})$ is time independent.

In the linear approximation [71, 72] (valid below and not far above the lasing threshold) we neglect the quadratic terms in (40) and use

$$\Delta n_{\omega}^{(0)} = 2\pi \, \Delta n_0(\mathbf{r}) \delta(\omega) \tag{41}$$

in (39). Polarization in this approximation is given by

$$\mathbf{P}_{\omega}^{(1)} = -\mathrm{i}(d^2/\hbar\gamma_{\perp})D(\omega)\Delta n_0(\mathbf{r})\mathbf{E}_{\omega} \tag{42}$$

and, when substituted into the right-hand side of (38), yields the following equation for the electric field:

$$-\epsilon(\mathbf{r}) \left(-v^2 + 2v\omega\right) \mathbf{E}_{\omega} + \nabla \times (\nabla \times \mathbf{E}_{\omega})$$

$$= -4\pi i \frac{d^2 v^2}{h \gamma_{\perp}} D(\omega) \Delta n_0(\mathbf{r}) \mathbf{E}_{\omega}, \tag{43}$$

where $D(\omega) \equiv [1 - i(\omega - v)/\gamma_{\perp}]^{-1}$.

One can use any of the system of modes $\psi_k(\omega; \mathbf{r})$ discussed in sections 2.2–2.4 and their adjoint modes $\phi_k(\omega; \mathbf{r})$ with respective eigenfrequencies $\Omega_k(\omega)$ (we use the same notation for all kinds of modes) in order to generate modal expansion (18) of the electric field in equation (43). The expansion coefficients are found using the biorthogonal functions as

$$a_k(\omega) = \int d\mathbf{r} \sqrt{\epsilon(\mathbf{r})} \, \boldsymbol{\phi}_k^*(\omega; \mathbf{r}) \cdot \mathbf{E}_{\omega}(\mathbf{r}). \tag{44}$$

Then (43) is reduced to the matrix eigenvalue problem

$$\sum_{k'} \left[\omega \delta_{kk'} - \bar{\Omega}_{kk'}(\omega)\right] a_{k'}(\omega) = 0 \tag{45}$$

with

$$\bar{\Omega}_{kk'}(\omega) = \Omega_k(\omega)\delta_{kk'} + i2\pi\nu \frac{d^2}{\hbar\gamma_\perp}D(\omega)V_{kk'}(\omega), \qquad (46)$$

$$V_{kk'}(\omega) = \int d\mathbf{r} \, \boldsymbol{\phi}_k^*(\omega; \mathbf{r}) \cdot \boldsymbol{\psi}_{k'}(\omega; \mathbf{r}) \frac{\Delta n_0(\mathbf{r})}{\epsilon(\mathbf{r})}.$$
 (47)

The matrix $V_{kk'}(\omega)$ becomes diagonal for uniform Δn_0 and ϵ . The lasing thresholds and frequencies at the thresholds are determined from the system of equations $\omega = \operatorname{Re} \bar{\Omega}_k(\omega)$ and $\operatorname{Im} \bar{\Omega}_k(\omega) = 0$, where $\bar{\Omega}_k(\omega)$ are eigenvalues of $\bar{\Omega}_{kk'}(\omega)$.

3.2. Third-order theory

Nonlinear effects can be included by iterating equations (39) and (40), with the field as a small parameter [64, 65]. Namely, $\mathbf{P}_{\omega}^{(1)}$ is inserted in (40) to obtain the correction $\Delta n_{\omega}^{(2)}$, which, in turn, is used in (39) to yield the contribution to the polarization $\mathbf{P}_{\omega}^{(3)}$, of the third order in the electric field.

Assuming that lasing modes exist, we can present the field as a sum of oscillating terms with slowly varying amplitudes,

$$\mathbf{E}(\mathbf{r},t) = \operatorname{Re}\left[\sum_{l} \mathbf{E}_{l}(\mathbf{r},t) e^{-i\omega_{l}t}\right],$$

$$\mathbf{E}_{\omega}(\mathbf{r}) = \sum_{l} \mathbf{E}_{l}(\omega - \omega_{l}; \mathbf{r}),$$
(48)

and positive frequencies ω_l close to ν , which are to be determined self-consistently. If the amplitudes $\mathbf{E}_l(\mathbf{r},t)$ vary slowly on the scale of ω_l , their Fourier transforms $\mathbf{E}_l(\omega-\omega_l;\mathbf{r})$ are strongly peaked at ω_l . The slowly varying amplitude approximation in the frequency domain amounts to the replacement $\mathbf{E}_l(\omega-\omega_l;\mathbf{r}) \to \pi \mathbf{E}_l(\mathbf{r},t) \, \delta(\omega-\omega_l)$ in nonlinear terms before performing frequency integrations when transforming to the time representation. Further, we average out interference terms, oscillating at the beat frequencies, and neglect mode degeneracies ($|\omega_l-\omega_m|\gg |\dot{\mathbf{E}}_l|/|\mathbf{E}_l|$). After the nonlinear correction $\mathbf{P}_\omega^{(3)}$ is added to the right-hand side of (38) and the linear contribution is diagonalized according to (45), we arrive at the third-order lasing equations

$$\left\{ \frac{\mathrm{d}}{\mathrm{d}t} + \mathrm{i}[\bar{\Omega}_{k}(\omega_{m}) - \omega_{m}] \right\} \bar{a}_{km}(t) = -\frac{\pi \nu}{\hbar \gamma_{\parallel}} \left(\frac{d^{2}}{\hbar \gamma_{\perp}} \right)^{2}
\times D(\omega_{m}) \int \mathrm{d}\mathbf{r} \frac{\Delta n_{0}(\mathbf{r})}{\sqrt{\epsilon(\mathbf{r})}} \bar{\boldsymbol{\phi}}_{k}^{*}(\omega_{m}; \mathbf{r}) \cdot \sum_{l=1}^{N_{m}} \left\{ 2 \operatorname{Re}[D(\omega_{l})] \right\}
\times \mathbf{E}_{m}(\mathbf{r}, t) |\mathbf{E}_{l}(\mathbf{r}, t)|^{2} + (1 - \delta_{ml}) D_{\parallel}(\omega_{m} - \omega_{l})
\times [D(\omega_{m}) + D^{*}(\omega_{l})] \mathbf{E}_{l}(\mathbf{r}, t) [\mathbf{E}_{l}^{*}(\mathbf{r}, t) \cdot \mathbf{E}_{m}(\mathbf{r}, t)] \right\}. (49)$$

 a_{km} is the amplitude of the *k*th component of the *m*th lasing mode in the basis of eigenfunctions of linearized problem (43) and (45):

$$\mathbf{E}_{m}(\mathbf{r},t) = \sum_{k=1}^{N_{b}} \bar{a}_{km}(t) \bar{\boldsymbol{\psi}}_{k}(\omega_{m};\mathbf{r}) / \sqrt{\epsilon(\mathbf{r})},$$

$$m = 1, \dots, N_{m}.$$
(50)

This is a system of $N_b \times N_m$ equations, where N_b is the basis size and N_m is the number of lasing modes. The terms proportional to $D_{\parallel}(\omega) \equiv (1 - i\omega/\gamma_{\parallel})^{-1}$ arise from the population pulsations at the beat frequency $\omega_m - \omega_l$ interfering in (39) with the oscillations at the frequency ω_l (and, thus, surviving the averaging). In the stationary regime ($d\bar{a}_{km}/dt = 0$), the amplitudes \bar{a}_{km} and frequencies ω_m can be determined from equations (49) by an iteration procedure (see section 3.3).

The number of equations in system (49) can be reduced to N_b if one assumes that the wavefunctions of the lasing modes are given by the linear approximation (43) and do not have to be determined self-consistently from the nonlinear equations (49). This is the approximation used in [71, 72], where it was justified by the fact that such a partially (in the

linear approximation) self-consistent treatment still allows one to eliminate fast oscillating terms in the nonlinear polarization and to introduce the slowly varying amplitude approximation resulting in rate equations for the respective amplitudes. In this linearly self-consistent approximation the amplitudes can be presented as $\bar{a}_{km}(t) = \bar{a}_m(t) \, \delta_{km}$ and the lasing equations for a scalar field take the form (cf [67, 69, 70])

$$\left\{ \frac{\mathrm{d}}{\mathrm{d}t} + \mathrm{i}[\bar{\Omega}_{m}(\omega_{m}) - \omega_{m}] \right\} \bar{a}_{m} = -\frac{\pi \nu}{V \hbar \gamma_{\parallel}} \left(\frac{d^{2}}{\hbar \gamma_{\perp}} \right)^{2}
\times D(\omega_{m}) \bar{a}_{m} \sum_{l} B_{ml} |\bar{a}_{l}|^{2} \times \left\{ 2 \operatorname{Re} \left[D(\omega_{l}) \right] + (1 - \delta_{ml}) \right.
\times D_{\parallel}(\omega_{m} - \omega_{l}) \left[D(\omega_{m}) + D^{*}(\omega_{l}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{m} - \omega_{l}) \left[D(\omega_{m}) + D^{*}(\omega_{l}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{m} - \omega_{l}) \left[D(\omega_{m}) + D^{*}(\omega_{l}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{m} - \omega_{l}) \left[D(\omega_{m}) + D^{*}(\omega_{l}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \left[D(\omega_{l}, \mathbf{r}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \left[D(\omega_{l}, \mathbf{r}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \left[D(\omega_{l}, \mathbf{r}) \right] \right\},$$

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$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \right\},$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right]$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right]$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right]$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right]$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right]$$

$$\left. \times D_{\parallel}(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right] \left[D(\omega_{l}, \mathbf{r}) \right]$$

where V is the volume. Equation (51) generalizes the standard third-order semiclassical theory with saturation (hole-burning) terms [64, 65] to the case of strongly open and irregular systems. Separating real and imaginary parts of this equation one obtains rate equations for intensities $I_m = |\bar{a}_m|^2$ of the modes and an equation for lasing frequencies:

$$\left\{ \frac{\mathrm{d}}{\mathrm{d}t} - 2\mathrm{Im}[\bar{\Omega}_{m}(\omega_{m})] \right\} I_{m} = -\frac{2\pi \nu}{V \hbar \gamma_{\parallel}} \left(\frac{d^{2}}{\hbar \gamma_{\perp}} \right)^{2} \\
\times I_{m} \operatorname{Re} \left[D(\omega_{m}) \sum_{l} (\cdots) \right], \tag{53}$$

$$\operatorname{Re}[\bar{\Omega}_{m}(\omega_{m})] - \omega_{m} = -\frac{\pi \nu}{V \hbar \gamma_{\parallel}} \left(\frac{d^{2}}{\hbar \gamma_{\perp}} \right)^{2} \\
\times \operatorname{Im} \left[D(\omega_{m}) \sum_{l} (\cdots) \right], \tag{54}$$

where the sum $\sum_{l} (\cdots)$ appearing in (51) depends on the intensities of all lasing modes. These rate equations do not contain any linear coupling terms, contrary to the assumption made in [66].

These equations show that all statistical characteristics of laser emission (frequency, threshold and intensity distributions) are determined by certain integrals involving eigenfunctions of cold cavities. The transition from strong to weak scattering manifests itself in changing statistical characteristics of the respective quantities. However, in spite of the large amount of work on wavefunction statistics in closed systems, the statistical properties of self- and cross-saturation coefficients in open resonators have not yet been studied. At the same time, it is clear now that this statistics is responsible for various regimes of behaviour of random lasers [70]. We will discuss this point in more detail in section 4.1.

3.3. All-order nonlinear theory in the time-independent population approximation

It is possible to obtain lasing equations valid in all orders in the electric field in a closed form if one neglects the time dependence of the population inversion. As seen from (49), the population-pulsation contribution can be neglected if $|D_{\parallel}(\omega_m - \omega_l)| \ll 1$. Typically, the lasing modes are excited

within the gain bandwidth γ_{\perp} around the atomic frequency. Then, the above condition reduces to $\gamma_{\perp} \gg \gamma_{\parallel}$.

Requiring that $\Delta n_{\omega}(\mathbf{r}) = \Delta n(\mathbf{r}) \, \delta(\omega)$, we express \mathbf{P}_{ω} from (39) and insert it in (40). If this assumption were actually consistent with equation (40) one would, after carrying out the mode-of-the-field expansion (48), end up (in the frequency representation) with terms which are also proportional to $\delta(\omega)$. In reality, in addition to 'correct' terms one would obtain a number of terms proportional to δ functions of various combinations of lasing frequencies, which describe oscillations of the population. Neglecting these 'oscillatory' terms is equivalent to keeping only diagonal contributions $|\mathbf{E}_l|^2$ in the modal expansion of the $\mathbf{E}^* \cdot \mathbf{P}$ term, quadratic in the field. In this approximation $\Delta n(\mathbf{r})$ can be determined self-consistently and inserted into (39) and (38) to obtain the lasing equations

$$\left\{ \frac{\mathrm{d}}{\mathrm{d}t} + \mathrm{i}[\Omega_{k}(\omega_{m}) - \omega_{m}] \right\} a_{km}(t)$$

$$= 2\pi \nu \frac{d^{2}}{\hbar \gamma_{\perp}} D(\omega_{m}) \int \mathrm{d}\mathbf{r} \frac{\Delta n_{0}(\mathbf{r})}{\sqrt{\epsilon(\mathbf{r})}}$$

$$\times \frac{\phi_{k}^{*}(\omega_{m}; \mathbf{r}) \cdot \mathbf{E}_{m}(\mathbf{r}, t)}{1 + \frac{d^{2}}{\hbar^{2} \gamma_{\perp} \gamma_{\parallel}} \sum_{l} \operatorname{Re}\left[D(\omega_{l})\right] |\mathbf{E}_{l}(\mathbf{r}, t)|^{2}}.$$
(55)

In contrast to the case for (49), the field here is expanded in the quasimodes of the passive system with the frequencies $\Omega_k(\omega_m)$, while the linear mode coupling is included in the right-hand side. These equations represent generalization of time-independent equations derived in [56], which are obtained from (55) by assuming time independence of the respective amplitudes. Equivalently, equation (55) can be derived by treating the polarization term in (38) as a source and using the Green function to write down the solution of this equation as [56]

$$\mathbf{E}_{\omega}(\mathbf{r}) = -\frac{4\pi v^{2}}{\sqrt{\epsilon(\mathbf{r})}} \int d\mathbf{r}' \, \epsilon^{-1/2}(\mathbf{r}') \, G(\omega; \mathbf{r}, \mathbf{r}') \, \mathbf{P}_{\omega}(\mathbf{r}')$$

$$= i \frac{4\pi v^{2}}{\sqrt{\epsilon(\mathbf{r})}} \frac{d^{2}}{\hbar \gamma_{\perp}} D(\omega) \int d\mathbf{r}' \, \frac{\Delta n_{0}(\mathbf{r}')}{\sqrt{\epsilon(\mathbf{r}')}}$$

$$\times \frac{G(\omega; \mathbf{r}, \mathbf{r}') \, \mathbf{E}_{\omega}(\mathbf{r}')}{1 + \frac{d^{2}}{\hbar^{2} \gamma_{\perp} \gamma_{\parallel}} \sum_{l} \text{Re} \left[D(\omega_{l}) \right] |\mathbf{E}_{l}(\mathbf{r}, t)|^{2}}.$$
(56)

Replacing the Green function with its spectral representation (22) in the rotating-wave approximation and integrating as in (44), we again arrive at equation (55). In the stationary case this equation is reduced to a nonlinear eigenmode problem

$$\sum_{k'} T_{kk'}(\omega_m) a_{k'm} = p^{-1} a_{km}, \tag{57}$$

$$T_{kk'}(\omega) = i2\pi \nu \frac{d^2}{\hbar \gamma_{\perp}} \frac{D(\omega)}{\omega - \Omega_k(\omega)} \int d\mathbf{r}' \frac{\delta n_0(\mathbf{r}')}{\epsilon(\mathbf{r}')} \times \frac{\boldsymbol{\phi}_k^*(\omega; \mathbf{r}') \cdot \boldsymbol{\psi}_{k'}(\omega; \mathbf{r}')}{1 + \frac{d^2}{\hbar^2 \nu_{\perp} \nu_0} \sum_{l} \text{Re} [D(\omega_l)] |\mathbf{E}_l(\mathbf{r})|^2}$$
(58)

where the unsaturated population inversion $\Delta n_0(\mathbf{r}) = p \, \delta n_0(\mathbf{r})$ is split into the overall pump strength p and the pump profile $\delta n_0(\mathbf{r})$. The field distribution in mode m is $\mathbf{E}_m(\mathbf{r}) = \sum_k a_{km} \psi_k(\omega_m; \mathbf{r})/\sqrt{\epsilon(\mathbf{r})}$. If the basis of constant-flux modes (section 2.4) is used, the field outside of the system can be obtained by continuation.

An algorithm for determining the lasing mode frequencies ω_m and expansion coefficients a_{km} as p is increased continuously from zero is described in [74]. Below the threshold, where all $a_{km} = 0$, one looks for the eigenvalues of the linear $T(\omega)$. Changing ω , the eigenvalues can be made real, one at a time. The largest real eigenvalue p_1^{-1} yields the threshold pump strength and the corresponding ω is the lasing frequency at the threshold. Above the threshold, for $p > p_1$, the pump is increased in small steps and the solution $a_{k1}(p)$ for the first mode is determined iteratively from (57). The second mode appears when the second-largest eigenvalue p_2^{-1} of $T(\omega)$ linearized 'around' the first mode becomes equal to p^{-1} . The procedure is continued to find higher modes.

4. Examples and properties of multimode random lasers

4.1. Threshold and number-of-modes statistics

The distribution of thresholds and the average number of lasing modes as a function of pump strength were calculated in [1] for an ensemble of weakly open chaotic cavities. Each cavity was opened via M small holes (diameter \ll wavelength), which together carry M open channels. The passive mode decay rates κ in this system are distributed according to the χ^2 distribution with M degrees of freedom,

$$P_M(y) = \frac{(M/2)^{M/2}}{\Gamma(M/2)} y^{M/2-1} \exp\left(-\frac{M}{2}y\right), \qquad y \equiv \kappa/\langle \kappa \rangle,$$
(59)

where $\Gamma(x)$ is the gamma function. The distribution is wide for small M and, for M=1, it increases as $y^{-1/2}$ when $y\to 0$. This property leads to a wide distribution of lasing thresholds, $\Delta n_{0,\text{thr}}$, which behaves as $\Delta n_{0,\text{thr}}^{M/2-1}$ for small $\Delta n_{0,\text{thr}}$. The average threshold is much less than the nominal value $\Delta \widetilde{n}_0 = \langle \kappa \rangle \hbar \gamma_\perp \epsilon / 2\pi \nu d^2$, which is the pumping required to overcome the average loss at $\omega = \nu$ (cf (46)). An increase of threshold fluctuations with localization was observed in a one-dimensional disordered model [42].

Considering the rate equation (53) in the stationary regime and neglecting the population-pulsation term containing $D_{\parallel}(\omega_m - \omega_l)$, one obtains a matrix equation for the intensities $I_m = |\bar{a}_m|^2$ of lasing modes:

$$\sum_{l} A_{ml} I_{l} = 1 - \frac{\Delta \tilde{n}_{0}}{\Delta n_{0}} \frac{y_{m}}{\mathcal{L}_{m}},$$

$$A_{ml} = \frac{2d^{2}V}{\epsilon \hbar^{2} \gamma_{\perp} \gamma_{\parallel}} \frac{\mathcal{L}_{1}}{\mathcal{L}_{m}} \text{Re} \left[D(\omega_{m}) B_{ml} \frac{\epsilon^{2}}{V^{2} \Delta n_{0}} \right],$$
(60)

where $y_m \equiv \kappa_m/\langle \kappa \rangle$ (κ_m is the decay rate of mode m) and $\mathcal{L}_m \equiv \text{Re } D(\omega_m)$. This equation must be complemented by the condition that it has only positive solutions. The rest of the basis are nonlasing modes and their intensities are set to zero. It should be noted, however, that the positiveness of lasing intensity does not, by itself, guarantee that the solution found is stable, a fact well known for simple two-mode models [64]. The stability of the solutions can only be verified from the time-dependent equation (53); therefore, the estimates of the

number of modes based on time-independent equations cannot, in general, be considered as completely accurate.

For a weakly open chaotic cavity [1] one can assume that (a) the eigenfunctions are almost real and orthogonal and (b) they can be described as random Gaussian functions which are uncorrelated for different modes [59]. If, in addition, one assumes that the background dielectric constant, $\epsilon(\mathbf{r})$, and pumping rate, $\Delta n_0(\mathbf{r})$, are both uniform, the correlator (52) takes the form of

$$B_{ml}\epsilon^2/V^2\Delta n_0 = 1 + 2\delta_{ml}. (61)$$

In this case matrix A_{ml} can be inverted analytically, yielding a dependence of the mode intensities on the pump strength. Let us order the modes $m=1,2,\ldots$ in the order in which they are excited as the pumping increases. It can be shown that, in this case, the y_m/\mathcal{L}_m form an increasing sequence. A threshold condition for the mode m results in the equation [1]

$$\left(\frac{m}{2}+1\right)\frac{y_m}{\mathcal{L}_m}-\frac{1}{2}\sum_{l=1}^m\frac{y_l}{\mathcal{L}_l}=\frac{\Delta n_0}{\Delta \widetilde{n}_0},\tag{62}$$

which relates the number of excited modes $N_{\rm m}=m$ and the pump strength Δn_0 in a particular cavity. The ensemble average $\langle N_{\rm m} \rangle$ is calculated [1] with the help of the distribution $P_M(y)$ for $y\ll 1$ and is found to scale as $\Delta n_0^{M/(M+2)}$ asymptotically for large pump strength.

In [67] $\langle N_{\rm m} \rangle$ was computed for nonweakly open cavities modelled by random matrices. In particular, the validity of the relation (61) was studied numerically for strong coupling to the bath $\gamma_n=1$ (section 2.5). It was shown that for $\kappa \lesssim \langle \kappa \rangle$ the assumption of uncorrelated eigenvectors works well, while for larger κ deviations from Gaussian statistics become stronger.

The power-law asymptotics for $\langle N_{\rm m} \rangle$ was confirmed by numerical simulation of decay rates entering (62) using random matrices with $\gamma_n \ll 1$ [68]. The standard deviation $\sigma_{N_{\rm m}}$ varied as $\Delta n_0^{M/2(M+2)}$. In the case $\gamma_n=1$, the ratio $\sigma_{N_{\rm m}}/\langle N_{\rm m} \rangle$, but not $\sigma_{N_{\rm m}}$ and $\langle N_{\rm m} \rangle$ separately, obeyed a power law with an exponent that depended on M. The difference between the results for the weak and strong couplings occurs because the decay rate distribution for $\gamma_n \sim 1$ is no longer of χ^2 type [58].

For a one-dimensional disordered system [70] assumption (61) fails completely, indicating a different type of mode competition compared to that for chaotic systems. Numerical simulations showed that the number of lasing modes saturates below the basis size with increasing pumping. This effect is related to the nonmonotonic dependence of mode intensities on the pump strength and complete disappearance of some modes for pumping exceeding certain thresholds.

The mode suppression was also reported for disordered disk lasers studied within the theory of section 3.3 [74]. Comparing the dependences of lasing frequencies and intensities on the pumping, it was noticed that when two modes come close together in frequency, one of them can be suppressed. The mode thresholds and intensities, but not the frequencies, were found to be very sensitive to the pumping spatial profile.

4.2. Frequency and intensity statistics

It is well known that passive closed chaotic systems without spatial symmetries have level repulsion, i.e., the probability density for zero frequency spacing vanishes. A natural question is that of how the spacing distribution for lasing modes in a random laser is connected to the distribution for passive modes in the underlying system without gain. It can be seen in the following examples that mode selection and competition normally enhance the repulsion in a laser. When lasing modes are close to passive modes, this property is rather obvious: even if two passive modes cross, the two of them will not necessarily lase.

Spacing distributions in two-mode chaotic lasers modelled with random matrices were computed numerically in [69]. Mode repulsion was present both in the case of weak openness and in that of intermediate openness, $\gamma_n \ll 1$ and $\gamma_n = 1$ (section 2.5), even though the passive frequencies can cross for $\gamma_n = 1$. (If two passive modes have the same frequencies, they have quite different lifetimes due to repulsion in the complex plane. Hence, only one of the two modes will be lasing for moderate pump strength.) When the gain bandwidth γ_{\perp} is close to the mean level spacing $\overline{\Delta \omega}$ in the passive system, the spacing distribution for the lasing modes is well described by the Wigner surmise [76]

$$P_{W}(\Delta\omega) = \frac{\pi}{2} \frac{\Delta\omega}{\overline{\Delta\omega}^{2}} \exp\left(-\frac{\pi}{4} \frac{\Delta\omega^{2}}{\overline{\Delta\omega}^{2}}\right), \tag{63}$$

derived for passive closed chaotic cavities in the random matrix theory. Again, this form works also for $\gamma_n = 1$, when the spacing statistics for passive modes is closer to Poissonian. This example shows that formal coincidence between a spacing distribution for lasing modes and the Wigner surmise does not guarantee that the physics behind them is the same.

Mode repulsion with deviations from the Wigner surmise was found numerically in one-dimensional disordered lasers [70]. There were two reasons for the repulsion. First, some modes were coupled, because the system was not very long and the modes could overlap. Second, when two modes were localized (and could have close frequencies), the mode that was closer to the opening had a higher threshold and was not excited.

When two modes have close frequencies, it may become necessary to take into account the dependence of mode frequencies on the pumping. As mentioned above, a correlation between mode repulsion and suppression during the change of the pumping level was observed in a numerical study of two-dimensional disordered laser [74].

Spacing distributions were measured in colloidal solutions containing TiO_2 scatterers and a laser dye [43]. The system was in the weak scattering regime in the sense that the scattering mean free path was much longer than the pump excitation cone. The lasing frequencies were more or less regularly spaced, exhibiting the mode repulsion. The spacing distribution had a maximum, but could not be fitted well with the Wigner surmise. The average mode spacing scaled with the dye concentration. (Increasing the concentration reduced the gain volume, which led to a reduction of the

number of modes.) The spacing fluctuations increased with the scatterer concentration. Some of the experimental results were supported by numerical simulations for a one-dimensional disordered system at the threshold. The statistics of lasing peaks was compared with the statistics of spontaneous-emission spikes that appear in the background of the emission spectra. The spikes were attributed to photons created in single spontaneous-emission events and amplified over long paths [41, 77]. Coherent feedback is not required for the appearance of spikes. The spikes' positions in the spectrum were uncorrelated, which was reflected in the Poisson spacing statistics.

Spectra with almost equally spaced lasing peaks were obtained for ${\rm TiO_2}$ colloidal solutions with strong reabsorption outside of the pumped volume [8]. Weak scattering, on the one hand, and reabsorption, on the other hand, result in an effective cavity being formed by just two scatterers located at the ends of the excitation cone. (The 'cavity' has the maximal possible length, because the gain grows exponentially with the path length, while the probability of photons leaving the cavity scales as a power of the length.) The effective cavity is of the Fabry–Perot type; therefore the lasing peaks are equidistant and the spacing scales inversely with the cone length.

Another system that shows mode repulsion is porous GaP filled with dye solution [3]. The transport mean free path was of the order of λ and about three times smaller than the pump spot size. The spacing distribution could be roughly fitted with the Wigner surmise.

The difference between lasing peaks and spontaneous-emission spikes for TiO_2 colloidal solutions with weak reabsorption [43] (see above) emerges also in the emission intensity statistics. Two statistical ensembles were considered: (1) intensities collected from all wavelengths in some range $I(\lambda)/\langle I(\lambda)\rangle$, normalized by the intensity averaged over many shots at given λ , and (2) peak and spike heights of the functions $I(\lambda)/\langle I(\lambda)\rangle$. For the two ensembles the probability distributions showed similar asymptotic behaviours at large intensities: they had a power-law tail above the lasing threshold and decayed exponentially below the threshold or in the absence of scatterers (neat dye solution). The numerically computed distribution of lasing mode intensities in a one-dimensional disordered laser [70] had a power-law decay, as well.

It should be mentioned that a power-law asymptotics may also appear for lasers with incoherent feedback near the threshold [78]. Thus, caution should be exercised when using an intensity distribution as a test for coherent lasing.

4.3. Structure of lasing modes

One of the important recent developments, which is relevant not only for random lasers, but also for the entire field of laser physics, is the realization of the fact that so called lasing modes may differ significantly from modes of passive cavities. It was noted in [71] that spatial nonuniformity of the refractive index and pumping can result in gain-induced linear coupling between modes of passive cavities, which results in the formation of new modes. These ideas were taken further

in the self-consistent theory of [56, 73–75], where no *a priori* assumptions about the spatial structure of lasing modes were made and they were found from the nonlinear self-consistent equation (55). Calculations of [56, 73–75] found significant modifications of the spatial profile of lasing modes due to nonlinear mode coupling. However, as we already mentioned, it is not clear whether the systems studied in [56, 73–75] can be considered as being in a diffusive regime. At the same time, no changes in the spatial structure of a lasing mode with increased pumping intensity were found in [27, 79], where the structures studied were clearly identified as being either in localized [27] or diffusive [79] regimes.

Modification of modes due to the presence of gain was observed in numerical simulations of a one-dimensional random laser below or at the threshold [42, 43], but only in the presence of spatially nonuniform (local) pumping. While modes in a uniformly pumped system were close to passive modes and their intensity grew exponentially towards the boundary, the transition to local pumping changed them substantially: they did not grow exponentially outside of the gain volume, but were still extended over the whole system. The number of lasing modes under local excitation was found to be less than the number of passive modes in the same frequency range, but larger than the number of passive modes in the reduced system defined by the pump region. In the case of nonuniform pumping the mode modification appears already in the linear approximation [72], so the roles of nonlinear effects in this simulation are unclear.

The structure of lasing modes was also studied in a system with weak scattering and strong reabsorption [8]. It was found that in such systems the gain volume surrounded by a strongly absorbing medium forms an effective cavity, where lasing modes are localized. Numerical simulations below and at the threshold in two dimensions showed that the lasing modes in this case are very close to the passive modes of the effective cavity.

5. Conclusions

Current research on multimode random lasing is moving along several major directions, e.g. (i) extension of conventional laser theories to open and irregular systems; (ii) statistical properties of lasing modes; (iii) mechanisms of random lasing (quantum and classical localization, extended modes), to name but a few. To date, a large number of experimental and numerical observations have been collected. However, the systems are hard to access analytically, as they consist of a number of strongly interacting components (electromagnetic field, gain medium, scatterers, boundaries) and several factors (openness, disorder, nonlinearity, noise) are not negligible over a wide range of parameters.

Recent important developments in the semiclassical multimode theory and random matrix theory added to the understanding of the properties of lasing modes in the stationary regime. To enable a direct comparison with experiments, mostly performed under pulsed-pumping conditions, it would be desirable to study time-dependent behaviour and relaxation processes. A detailed analysis

of mode stability, hysteresis phenomena and quantum-noise effects in the stationary regime is also lacking. To address the role of localization in the lasing feedback at an adequate level, numerical simulations of more realistic (three-dimensional) models might be necessary.

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