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Statistical theory of multimode random lasers

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Abstract
We present a semiclassical laser theory for multimode lasing in optical resonators with overlapping modes. Nonlinear saturation and mode competition are characterized in terms of left and right eigenmodes of a non-Hermitian operator. The theory is applied to wave-chaotic cavities and weakly disordered random media. In the limit of sufficiently strong pumping, we find that the mean number of laser peaks increases with the $1/3$ power of the pump strength.

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In recent years random laser action has been demonstrated [1–3] in a variety of disordered dielectric media. Feedback in these media is supplied by the disorder-induced random scattering of light. Lasing is seen in a drastic spectral narrowing and the appearance of discrete peaks in the emission spectrum above the laser threshold. The photon statistics within the peaks approaches a Poissonian distribution typical for coherent laser light.

While there is by now a substantial literature on random lasers [3], one of their most interesting features has not been fully understood: the strong increase of the number of laser peaks with increasing pump intensity. Numerical investigations of the time-dependent laser equations [4–6] find a saturation of the peak number at a value proportional to the system size, but these studies address the regime of spatially localized modes in one- and two-dimensional systems. The rate equations of Misirpashaev and Beenakker [7] predict a power-law increase of the peak number with pump intensity, but these equations are based on standard multimode laser theory [8] that is restricted to spectrally isolated resonator modes. Random laser modes, however, are generically weakly confined and spectrally overlapping in the regime of weak pumping.

In the present paper, we address the number of laser peaks in random media by generalizing multimode laser theory to weakly confining resonators with overlapping modes. We derive semiclassical laser equations that take into account both self-saturation and the mode competition of several laser modes for the atomic gain. Combining the laser equations with random-matrix theory, we compute the number of laser peaks for wave-chaotic and disordered media in the diffusive regime. The mean number of laser peaks $\langle N\rangle$, averaged over appropriate random-matrix ensembles, shows a universal increase as a function of the pump intensity S. For sufficiently strong pump, we find $\langle N\rangle \sim (S/S_0)^{1/3}$ (S_0 is a reference pump value) where the exponent $1/3$ is only determined by the wave-chaotic nature of the random laser modes and by mode competition.

We start with the formulation of the multimode laser theory in a general form and then discuss the application to random media. We consider an optical resonator filled with an active medium represented by a large number of homogeneously broadened two-level atoms. The atomic transition frequency ν is assumed to be much larger than the level broadening $\gamma_\perp$. The resonator is non-ideal due to leakage into the external electromagnetic field. We allow for resonator losses comparable or larger than the mode separation resulting in strongly overlapping resonator modes. The atom–field dynamics is described by the nonlinear equations of quantum laser theory

$$\dot{b}_\lambda = -\mathrm{i}\omega_\lambda b_\lambda - \pi \sum_{\lambda'} (VV^\dagger)_{\lambda\lambda'} b_{\lambda'} + \int \mathrm{d}\mathbf{r}\, g^*_\lambda(\mathbf{r})\sigma_-(\mathbf{r}) + F_\lambda, \tag{1}$$

$$\dot{\sigma}_-(\mathbf{r}) = (-\mathrm{i}\nu - \gamma_\perp)\sigma_-(\mathbf{r}) + 2\sum_\lambda g_\lambda(\mathbf{r})\sigma_z(\mathbf{r}) b_\lambda + F_-(\mathbf{r}), \tag{2}$$

$$\dot{\sigma}_z(\mathbf{r}) = \gamma_\parallel (S\rho(\mathbf{r}) - \sigma_z(\mathbf{r})) - \sum_\lambda (g^*_\lambda(\mathbf{r}) b^*_\lambda \sigma_-(\mathbf{r}) + \mathrm{c.c.}) + F_z(\mathbf{r}). \tag{3}$$

Here, the operator b_λ represents the field amplitude in the resonator mode λ and $\rho(\mathbf{r})$ the density of atoms. The operators $\sigma_-(\mathbf{r})$ and $\sigma_z(\mathbf{r})$ denote the atomic polarization and inversion density averaged over all atoms within a volume of size less than an optical wavelength, centred around the point $\mathbf{r}$. The decay rates $\gamma_\perp$, $\gamma_\parallel$ and the pump intensity S result from the interaction between atoms and external baths. That interaction gives rise to the noise forces $F_-(\mathbf{r})$, $F_z(\mathbf{r})$, while the noise term F_λ is induced by the external radiation field. The field–atom coupling constants are given by $g_\lambda(\mathbf{r}) = \nu d\phi_\lambda(\mathbf{r})/\sqrt{2\hbar\epsilon_0\omega_\lambda}$ where d is the atomic dipole matrix element. The functions ϕ_λ form a complete, orthonormal set of resonator eigenstates. We assume all noises to be Gaussian distributed and short-time correlated.

Our laser equations differ in one important aspect from the equations of standard laser theory: standard theory [8] models the passive resonator modes by independent oscillators, while the field amplitudes in equation (1) are coupled by the damping matrix $VV^\dagger$. The damping originates from the coupling between the resonator modes and the external electromagnetic field [9, 10]. Standard theory is recovered for weak coupling upon keeping only the diagonal elements of $VV^\dagger$.

Various limiting cases of the above equations have been studied before. For random media in the weak pumping limit, mode overlap gives rise to an excess noise of amplified spontaneous emission discovered by Beenakker [10–12]. The linewidth of a single lasing oscillation is enhanced by the Petermann excess noise factor [9, 13–15]. Lasing with nonresonant feedback [16, 17] is described by rate equations that follow from equations (1)–(3) assuming a number of overlapping laser modes above threshold. With increasing pump strength the spectral width of these laser oscillations decreases, ultimately yielding a spectrum of few or many quasi-discrete laser lines. It is this regime of multimode lasing that we explore in the present paper.

Semiclassical laser theory amounts to neglecting all noise terms in equations (1)–(3) and to replacing operators by their c-number expectation values. For simplicity we stick to the notation introduced before, and use b_λ, $\rho(\mathbf{r})$, $\sigma_-(\mathbf{r})$ and $\sigma_z(\mathbf{r})$ to denote the respective mean values. We restrict ourselves to laser media for which the characteristic times for atomic pump and losses are short compared to the lifetimes of photons in the resonator. Then one can adiabatically eliminate the atomic variables and derive a set of nonlinear equations for the field amplitudes alone.

Insight into this nonlinear problem is obtained starting from the weak pumping regime. In that regime one can linearize the laser dynamics by setting $\sigma_z(\mathbf{r}) \equiv S\rho(\mathbf{r})$. The (only) stationary solution has zero field amplitude, $\bar{b}_\lambda = 0$, in all modes. Deviations from the stationary state relax to zero with characteristic complex frequencies ω_k that follow from linear stability analysis: substituting the ansatz $\delta b_\lambda(t) = \delta b_\lambda \exp(-\mathrm{i}\omega t)$ for all modes λ into the equations-of-motion one obtains the secular equations

$$\sum_{\lambda'}[\omega\delta_{\lambda\lambda'} - \mathcal{H}_{\lambda\lambda'}]\delta b_{\lambda'} = 0, \qquad \lambda = 1, 2, 3, \ldots. \tag{4}$$

They imply that the frequencies ω_k are the eigenvalues of the non-Hermitian matrix $\mathcal{H}$ with the matrix elements

$$\mathcal{H}_{\lambda\lambda'} = \omega_\lambda\delta_{\lambda\lambda'} - \mathrm{i}\pi(VV^\dagger)_{\lambda\lambda'} + \mathrm{i}G_{\lambda\lambda'}(\omega), \tag{5}$$

$$G_{\lambda\lambda'}(\omega) \equiv 2S\int \mathrm{d}\mathbf{r}\rho(\mathbf{r})\frac{g^*_\lambda(\mathbf{r})g_{\lambda'}(\mathbf{r})}{\mathrm{i}(\nu-\omega)+\gamma_\perp}. \tag{6}$$

The three terms on the right-hand side of equation (5) account, respectively, for the deterministic internal resonator dynamics, the escape losses and the linear gain. In general, both the eigenvalues and the eigenvectors of $\mathcal{H}$ depend parametrically on the pump strength S. A simplification arises when the atoms are uniformly distributed over the resonator volume $\mathcal{V}$ with density $\rho(\mathbf{r}) = \mathcal{N}/\mathcal{V}$. Then the integration in equation (6) can be performed. Approximating the field–atom couplings $g_\lambda(\mathbf{r}) \approx d\Phi_\lambda(\mathbf{r})\sqrt{\nu/2\hbar\epsilon_0}$ this yields the gain $G_{\lambda\lambda'}(\omega) = G(\omega)\delta_{\lambda\lambda'}$, where

$$G(\omega) = 2S\frac{\mathcal{N}g^2}{\mathrm{i}(\nu-\omega)+\gamma_\perp}, \tag{7}$$

and $g \equiv d\sqrt{\nu/2\hbar\epsilon_0\mathcal{V}}$. Combination of equations (5) and (7) shows that the eigenvectors of $\mathcal{H}$ are independent of S. The eigenvalues are simply related to the eigenvalues $\Omega_k - \mathrm{i}\kappa_k$ of the passive resonator, yielding the spectral decomposition

$$\mathcal{H} = \sum_k(\Omega_k - \mathrm{i}\kappa_k + \mathrm{i}G_k)|R_k\rangle\langle L_k|. \tag{8}$$

Here, $\langle L_k|$ and $|R_k\rangle$ are the left and right eigenvectors of the passive cavity, and $G_k = G(\omega_k)$ is the linear gain at frequency ω_k determined self-consistently from the relation $\omega_k = \Omega_k - \mathrm{i}\kappa_k + \mathrm{i}G_k$. Generically, the eigenvectors form a complete bi-orthogonal set. They can be normalized to satisfy $\langle L_k|R_l\rangle = \delta_{kl}$. Upon increasing the pump intensity S, all eigenvalues move up in the complex plane. The laser threshold is reached when for the first time the loss rate κ_k of an eigenvalue equals its gain $\mathrm{Re}\, G_k$. Then the trivial solution $\bar{b}_k = 0$ becomes unstable and the non-zero laser field amplitudes must be determined from the nonlinear laser equations.

The multimode equations can now be derived generalizing methods from standard laser theory [8]. Several modes with indices k may perform laser action. For these modes, we make the ansatz

$$b_\lambda(t) = \sum_k\langle\phi_\lambda|R_k\rangle\beta_k(t), \qquad \beta_k(t) = \bar{b}_k(t)\,\mathrm{e}^{-\mathrm{i}\bar{\Omega}_k t}, \tag{9}$$

with yet unknown positive frequencies $\bar{\Omega}_k$ and complex amplitudes $\bar{b}_k$. We admit the $\bar{b}_k$'s to be weakly time dependent but that time evolution shall take place on time scales much larger than the oscillation periods $1/\bar{\Omega}_k$. We confine our analysis to laser modes not too far above threshold, so that the field intensities are not too large. Specifically, we assume that $\epsilon = g^2|\beta|^2/(\gamma_\perp\gamma_\parallel) \ll 1$ where $|\beta|^2$ denotes a typical field intensity. Then one may expand the laser equations keeping only terms up to third order in the field amplitudes. This yields

$$\frac{\mathrm{d}\beta_k}{\mathrm{d}t} = (-\mathrm{i}\Omega_k - \kappa_k + G_k)\beta_k - 4S\frac{\mathcal{N}}{V}\sum_{k_1,k_2,k_3} M_{kk_1k_2k_3}\int \mathrm{d}\mathbf{r}\, g_k^{L*}(\mathbf{r})g_{k_1}^R(\mathbf{r})g_{k_2}^{R*}(\mathbf{r})g_{k_3}^R(\mathbf{r})\beta_{k_1}\beta_{k_2}^*\beta_{k_3}. \tag{10}$$

These equations generalize the multimode equations of standard laser theory to resonators with overlapping modes. For each laser oscillation there are two coupling amplitudes

$$g_k^L(\mathbf{r}) = g\sqrt{\mathcal{V}}L_k(\mathbf{r}), \qquad g_k^R(\mathbf{r}) = g\sqrt{\mathcal{V}}R_k(\mathbf{r}) \tag{11}$$

to the left and right modes, L_k and R_k, respectively. Standard laser theory is recovered in the limiting case when the coupling between the resonator and the external radiation field is weak. Then left and right modes for each k become identical up to a trivial normalization factor, and g_k^L and g_k^R coalesce to the single coupling amplitude g_k between atoms and an eigenstate of the *closed* resonator. The coefficient $M_{kk_1k_2k_3}$ is a function of the atomic rates $\gamma_\perp, \gamma_\parallel$, the transition frequency ν and the laser frequencies $\bar{\Omega}_k, \ldots, \bar{\Omega}_{k_3}$, and takes the same form as in standard multimode theory [8].

Equations (10) admit different types of solutions depending on whether phase relations between laser oscillations exist or not. We focus on the so-called normal or free-running case when different laser modes oscillate independently from each other. The phases of these oscillations are uncoupled. In this case, one can multiply equation (10) by $\langle R_k|R_k\rangle\beta_k^*$ and average both sides of the resulting equation over time intervals much larger than the oscillation periods. Equivalently, one may perform a phase average assuming that the phases of different laser oscillations are uncorrelated. Taking real and imaginary parts, one obtains two sets of equations

$$0 = (-\kappa_k + \mathrm{Re}\, G_k)I_k - \sum_l (\mathrm{Re}\,\mathcal{B}_{kl})I_k I_l, \tag{12}$$

$$\bar{\Omega}_k = \Omega_k - \mathrm{Im}\, G_k + \sum_l (\mathrm{Im}\,\mathcal{B}_{kl})I_l, \tag{13}$$

from which one can determine the laser frequencies $\bar{\Omega}_k$ and the total field intensities $I_k = \langle R_k|R_k\rangle|\bar{b}_k|^2$ integrated over the resonator volume. The parameter $\mathcal{B}_{kl}$ characterizes the nonlinearity and is given in terms of the left and right resonator modes

$$\mathcal{B}_{kl} = 4S\mathcal{N}g^4[(\mathcal{M}_{kkll} + \mathcal{M}_{kllk})F_{kl}], \tag{14}$$

$$F_{kl} = \frac{\mathcal{V}\int_{\mathcal{V}} \mathrm{d}\boldsymbol{r}\, L_k^*(\boldsymbol{r})R_k(\boldsymbol{r})R_l^*(\boldsymbol{r})R_l(\boldsymbol{r})}{\langle L_k|R_k\rangle\langle R_l|R_l\rangle}. \tag{15}$$

In the weak-coupling case, it is easily checked that the nonlinearity reduces to the well-known expression of standard theory involving a fourth-order product of closed-resonator eigenstates.

We now turn to the application of our results to random media. As a first example, we compute the number of lasing modes in wave-chaotic resonators defined through irregularly shaped mirrors. One or several openings provide for a coupling to the external radiation field. Note that the passive resonator is then in the regime of overlapping modes. The corresponding laser problem is therefore fundamentally different from the problem of lasing with spectrally isolated modes considered in [7]. We address the problem using the rate equation approximation to equations (12), (13), i.e. we identify the laser frequencies $\bar{\Omega}_k$ with the passive resonator frequencies Ω_k and calculate the field intensities from

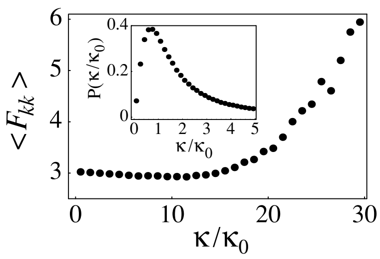

Figure 1. The wavefunction correlator averaged over a large ensemble of non-Hermitian random matrices $\mathcal{H}_{\rm eff}$ supporting $M = 5$ open channels as a function of the decay rate κ/κ_0. Inset: distribution of decay rates for the same random-matrix ensemble.

equation (12). As a consequence of the chaotic scattering at the resonator mirrors both the loss rates κ_k and the nonlinearities $\mathcal{B}_{kl}$ become random quantities. Their statistics can be obtained from the random-matrix model of chaotic scattering [18, 19]: one represents the passive resonator dynamics by large non-Hermitian random matrices $\mathcal{H}_{\rm eff} = H - {\rm i}\pi V V^\dagger$ where H is an $L \times L$ matrix taken from the Gaussian orthogonal ensemble with probability density $P(H)dH \propto \exp[-L\,{\rm Tr}\,H^2]dH$. The $L \times M$ rectangular matrix W describes the coupling to M fully transmitting channels. The distribution of decay rates has been calculated in [19]. In the limiting cases of small, respectively large, rates the result is

$$P(\kappa) \sim \begin{cases} \kappa^{M/2-1} & \text{for} \quad \kappa \ll \kappa_0 \\ \kappa^{-2} & \text{for} \quad \kappa \gg \kappa_0 \end{cases} \tag{16}$$

where $\kappa_0 = M\Delta/4\pi$ is a typical decay rate and Δ is the mean spacing of neighbouring eigenfrequencies of H.

Much less is known about the *eigenvector* statistics of non-Hermitian matrices [20]. Schomerus *et al* [21] showed that this statistics is directly related to the statistics of the laser linewidth in single mode random lasers. However, the correlation function (15) that enters our multimode laser equations has not been considered before. We have studied that correlator numerically doing random-matrix simulations. Figure 1 shows the diagonal correlator $\langle F_{kk}\rangle$ computed for a given value of κ and $M = 5$ open channels, averaged over a large number of non-Hermitian random matrices. One observes $\langle F_{kk}\rangle \approx 3$ for all resonances with decay rates less than or comparable to κ_0; larger eigenvector correlations are found for very broad resonances when $\kappa \gg \kappa_0$. The value $\langle F_{kk}\rangle = 3$ is reproduced upon simply counting contractions, assuming that the eigenvector components of L_k and R_k are independent Gaussian random variables only restricted by the requirements $\langle L_k|R_k\rangle = 1$ and $\langle R_k|R_k\rangle = 1$. The large correlations visible in figure 1 therefore indicate deviations from Gaussian statistics for eigenvectors associated with very broad resonances. However, the resonances relevant for lasing generally have width smaller than or comparable to κ_0. The associated eigenvector correlations are self-averaging in the limit of large matrices $L \to \infty$ and can be approximated by $\langle F_{kl}\rangle = 1 + 2\delta_{kl}$. Substitution into equation (14) and combination with equation (12) yields the rate equation

$$\left(-\kappa_k + SW_k - 2\frac{S}{\gamma_\parallel}\sum_l W_k W_l (1 + 2\delta_{kl}) I_l\right) I_k = 0, \tag{17}$$

where we introduced the rate $W_k = (1/S)\,{\rm Re}\,G_k$ for decay of excited atoms into the mode k. The three terms on the left-hand side account, respectively, for the escape loss, the linear gain

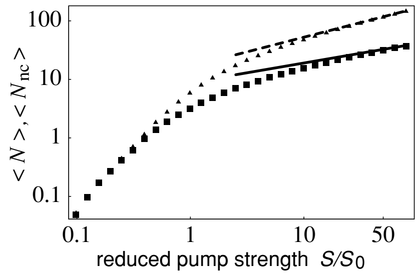

Figure 2. Average number of lasing modes $\langle N \rangle$ (squares) versus reduced pump strength S/S_0. The triangles represent the average number $\langle N_{\rm nc} \rangle$ of non-competing modes whose linear gain exceeds their loss. The solid and the dashed lines are the power-law estimates based on the continuum approximation to equation (18).

and the nonlinearities. Random scattering enters equation (17) through the random-matrix statistics of the decay rates κ_k.

Equations of the type (17) have been considered before, in the context of essentially one-dimensional ring lasers [8] and of wave-chaotic laser cavities with almost totally reflecting mirrors [7]. Our problem differs from [7] in that we allow for spectrally overlapping modes in the passive system. The mode correlator $\langle F_{kl} \rangle$ turns out not to be affected by the mode overlap (except for modes with very large decay rates). However, the statistics of the mode decay rates κ_k is substantially different. We note that the results of [7] can be recovered as a special case of our general theory in the limit of spectrally isolated modes.

Applying the rate equation method developed in [8], we can determine the number of lasing modes: first one labels the modes in order of increasing loss–gain ratio, so that $\kappa_1/W_1 \leqslant \kappa_2/W_2 \leqslant \cdots \leqslant \kappa_L/W_L$. The number N of lasing modes is restricted by the requirement that all intensities I_k with $k = 1, 2, \ldots, N$ must be positive. This yields the condition [7]

$$(2+N)\frac{\kappa_N}{W_N} - \sum_{k=1}^{N} \frac{\kappa_k}{W_k} < 2. \qquad (18)$$

The maximum integer N that fulfils this condition is the number of lasing modes. Its mean value is found using equation (18) for a given realization of the matrix $\mathcal{H}_{\rm eff}$ and then performing the random-matrix average. In figure 2, we compare $\langle N \rangle$ (squares) with the mean number of modes $\langle N_{\rm nc} \rangle$ (triangles) whose linear gain exceeds their losses; in other words, $N_{\rm nc}$ counts the number of modes that would be lasing if there was no mode competition. Both averages are shown as a function of the dimensionless pump strength S/S_0, where S_0 is the value of pump for which the linear gain at frequency $\Omega = \nu$ equals the typical loss κ_0 (from equation (7) $S_0 = \kappa_0\gamma_\perp/(2\mathcal{N}g^2)$). Clearly $\langle N \rangle \leqslant \langle N_{\rm nc} \rangle$ since mode competition can only reduce the number of lasing modes. In the limit of both small and large pumps one observes a power-law increase $\langle N \rangle \sim (S/S_0)^u$. The exponent u can be found combining the continuum approximation to equation (18) with the asymptotic form of the width distribution (16). For small S this argument yields $u = M/2$ for both $\langle N \rangle$ and $\langle N_{\rm nc} \rangle$. In the opposite limit of large pumping, the number of lasing modes grows with the power $u = 1/3$ while $u = 1/2$ is found in the absence of mode competition. We note that these exponents are *universal*: they neither depend on the shape of the mirrors nor on the number of open channels, provided this number is sufficiently large to ensure that the (passive) resonator modes are overlapping. The origin for this large degree of universality is the combination of mode overlap and chaotic scattering with mode competition[1].

A second example is disordered media in the diffusive regime. Eigenmodes in that regime are generically extended and exhibit correlations similar to those found in chaotic cavities. As a consequence, the mean number of lasing modes is expected to increase with the same power $1/3$ as found before for sufficiently large pump values. The above picture, however, does not apply to anomalously localized or prelocalized modes [22]. Such modes have anomalously high quality factors and unusual correlations not described by random-matrix theory. The statistics of laser peaks induced by these modes is an open problem.

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