Effects of spatial non-uniformity on laser dynamics.

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Semiclassical equations of lasing dynamics are re-derived for a lasing medium in a cavity with a spatially non-uniform dielectric constant. The non-uniformity causes a radiative coupling between modes of the empty cavity, which results in a renormalization of self- and cross-saturation coefficients. Possible manifestations of these effects in random lasers are discussed.

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Introduction  Random lasers, in which optical feedback is provided by scattering of light due to spatial inhomogeneity of the medium rather than by well defined mirrors, has recently attracted a great deal of attention.

In the case of weak scattering, when the propagation of light can be described within diffusion approximation, the nature of lasing in such systems has been well understood starting with a pioneering work by Letokhov and followed by a large volume of subsequent experimental and theoretical studies. The case of strong scattering, however, when light can be at the verge of Anderson localization, remains much more controversial. Experimental results of Ref.9 and consecutive works with strongly scattering systems (see recent reviews in Ref.1 and Ref.2) led to an assumption that lasing observed in those experiments is due to formation of pre-localized, if not completely localized states of light, which play a role of lasing cavities, and provide coherent resonant optical feedback as oppose to non-resonant feedback affecting only intensity of light in the diffusion case. The presence of narrow multiple lasing peaks4 as well as Poisson statistics of emitted radiation10 were considered as evidences in the favor of this interpretation of these experiments. However, it was shown in Ref.11 that the non-resonant feedback can also result in lasing with multiple narrow peaks. Moreover, the authors of Ref.12 demonstrated that the Poisson statistics also cannot be considered as an exclusive attribute of lasing with the resonant feedback.

In this situation, the recent results of Ref13 assume a particular significance. In these experiments a multipeak lasing was observed in PMMA sheets containing a rhodamine dye as an active material and titanium dioxide microparticles as scatterers. This system is characterized by a strong inhomogeneous broadening of the lasing transition, and most of the lasing peaks are separated in the frequency domain by a homogeneous line width of the lasing transition, $\gamma_a$. This is naturally explained by the competition of modes “feeding” from the same population inversion and spectral hole burning in inhomogeneously broadened systems13. However, above certain value of the pumping intensity, there were observed two lasing peaks coexisting within the homogeneous line width, $\gamma_a$ and having synchronized temporal behavior. This observation is indicative of the genuine two-mode lasing, which can occur in regular cavity lasers, when the mode competition is weakened by spatial hole burning10. Such a behavior, however, cannot take place in the case of the non-resonant feedback, because, in diffusive systems lasing only occur at the frequency of an atomic transition (multiple peaks in Ref.7 are due to inhomogeneous broadening of the transition used to generate emission and do not signify a truly multi-mode behavior).

Thus, as of today, the results of Ref13 provide the most convincing evidence of the resonant feedback in random lasers. It is important, therefore, to achieve a clear understanding of the specifics of non-linear mode interaction in such systems. However, since the experiments of Ref13 deal with just a single realization, the randomness, by itself, is not important here. What is important is the spatial inhomogeneity of the quasi-cavity, supporting the modes of interest. The main objective of this paper, therefore, is to study how this spatial inhomogeneity affects lasing threshold and spatial hole burning. Our consideration, however, is not constrained by random lasers, and can be applied to any type of lasers with spatially non-uniform cavities. Currently, there is a tremendous interest to lasing in the systems with a modulated dielectric constant, for instance, photonic crystals. The result presented here are relevant for these systems as well. Moreover, current technologies allow for engineering structures with virtually arbitrary spatial profile of the dielectric function. The results of this paper can be used to manipulate properties of lasers by using spatial dependence of the cavity dielectric function as a new design parameter.

A general multi-mode theory of lasing in systems with an arbitrarily inhomogeneous dielectric constant, $\epsilon(r)$, presented here is an extension of semi-classical Lamb theory10 for the media whose dielectric constant is inhomogeneous in the direction of propagation of the laser beam (inhomogeneity in the perpendicular directions results in waveguiding effects, which are well studied in laser physics (see, for instance,11)). This inhomogeneity modifies the orthonormalization condition for the eigen modes of the cavity, making the standard inner product of the modes belonging to different eigen frequencies dif-
different from zero. The main effect resulting from this non-orthogonality is a new type of linear coupling between normal modes of the empty cavity, which is mediated by the polarization of the active medium.

The non-orthogonality of eigen modes due to the inhomogeneity of $\epsilon(r)$ should not be confused with non-orthogonality of Fox-Li modes of uniform but leaky (open) cavities, which arises due to non-hermitian nature of the respective eigen value problem and does not result in any additional linear coupling between the modes. The only consequence of the non-hermitian nature of such cavities is the presence of an additional factor in the linear susceptibility of the active medium, which was carefully studied in the past and shown to be responsible for the excess noise in unstable cavities.

Multi-mode laser equations for an inhomogeneous medium We consider an ideal cavity specified by an inhomogeneous dielectric function $\epsilon(r)$ and some boundary conditions. The cavity is filled with an active medium characterized by its polarization $\mathbf{P}(r)$. Let us assume that we know the full system of eigen modes, $f_k(r)$, and respective eigen frequencies $\omega_k$ of such a cavity in the absence of the polarization. These modes can be used to present electric field, $E$, and polarization $P$ in the form of their linear combinations: $E = \sum_k E_k(t) f_k(r)$, $P = \sum_k P_k(t) f_k(r)$, where we assume that only s-polarized modes couple to the active medium, and ignore the vector nature of the field and the polarization. The orthonormalization condition for these modes involves inhomogeneous dielectric function $\epsilon(r)$: $\int \epsilon(r) f_k^*(r) f_{k'}(r) dr = \delta_{kk'}$, which means that the wave functions $f_k(r)$ themselves are neither normalized nor orthogonal. As a result, the dynamic equations for the amplitudes, $E_k$, takes the following form

$$\ddot{E}_k(t) + (\omega_k - i\gamma_k)^2 E_k(t) = -4\pi \sum_{k_1} V_{kk_1} \dot{P}_{k_1}(t), \quad (1)$$

where we introduced cavity losses, characterized by phenomenological parameters $\gamma_k$. The main peculiarity of Eq. (1) is the presence of the linear coupling between different polarization amplitudes, $P_k$, characterized by non-diagonal elements of the matrix

$$V_{kk_1} = \int f_k^*(r) f_{k_1}(r) dr \quad (2)$$

The presence of such a coupling is the main difference between homogeneous and inhomogeneous cavities. The magnitude of coupling parameters $V_{kk_1}$ depends on the spatial profile of the dielectric constant, and can be tailored to enhance (or diminish) the coupling effects.

Similar equation can be in principle derived for inhomogeneous open cavities as well, where eigen modes $f_k$ should be replaced by appropriate Fox-Li modes. The hermitian orthogonality in this case is replaced by the bi-orthogonality, which involves adjoint set of modes.

A general case of non-uniform open cavity this condition was derived, for instance, in Ref. 14. We shall leave, however, this topic for future work.

A gain medium is described within the model of two-level atoms, characterized by dephasing rate $\gamma_a^{-1}$, and population relaxation time $\tau$, and we use a standard density matrix approach in order to derive equations for polarization amplitudes $P_k$, and population difference $\Delta N$. The next standard step in the derivation of rate equations would be rotating wave and slow amplitude approximations, which amount to presenting mode amplitudes $E_k$ and $P_k$ as $E_k(t) = \Xi_k(t) \exp(-i\Omega_k t)$, $P_k(t) = \Pi_k(t) \exp(-i\Omega_k t)$, where $\Xi_k$ and $\Pi_k$ are slowly changing amplitudes, and $\Omega_k$ is a frequency of the respective lasing mode. However, forcing this procedure onto equation (1) yields linear oscillating terms of the form $\sum V_{kk_1} \chi(1)(\Omega_{k_1}) \exp[-i(\Omega_k - \Omega_{k_1})] \Xi_{k_1}$, which render derivation of meaningful rate equations impossible.

Here

$$\chi(1)(\omega) = \frac{|\mu|^2 \Delta N_0}{4\hbar} \frac{1}{\omega - \omega_0 + i\gamma_a} \quad (3)$$

is a linear susceptibility of the gain medium with $|\mu|$ being dipole matrix element of the lasing transition. Parameter $\Delta N_0$ represents non-saturated population inversion, and characterizes the strength of the pumping.

The physical origin of this problem is quite clear — in the presence of the linear coupling the modes of a passive medium are not genuine normal modes of the entire system. As a result, an attempt to excite such a mode leads to exchange of energy between coupled modes and to non-stationary oscillations of the respective intensities. The rate equations, therefore, should be derived for the normal modes of the entire system, which would include cavity and the gain medium. To this end, it is convenient to transform Eq. (1) in the frequency domain using conventionally defined Fourier transformation:

$$\sum_k \left[ (\omega_k - i\gamma_k - \omega) \delta_{kk_1} - 2\pi \omega_0 V_{kk_1} \chi(1)(\omega) \right] \tilde{E}_{k_1}(\omega) = 2\pi \omega_0 \sum_k V_{kk_1} \tilde{P}^{(3)}_{k_1}(\omega) \quad (4)$$

where a tilde on the top of a symbol signifies the Fourier transform of the respective quantity, and the polarization is separated into a linear and third-order non-linear contribution, $\tilde{P}^{(3)}$. The former is taken into account in Eq. (1) by introducing a linear susceptibility $\chi(\omega)$, and the expression for the latter was derived in a standard way from the full system of density matrix equation using standard perturbation approach.

$$\tilde{P}^{(3)}_k = \frac{|\mu|^2 \Delta N_0}{32\pi^2 \hbar^3} \sum_{kk_1kk_2k_3} A_{kk_1kk_2k_3} \int d\omega_1 d\omega_2 \times (5)$$

$$\tilde{E}_{k_1}(\omega - \omega_1) \left[ \frac{\tilde{E}_{k_2}(\omega_2) \tilde{E}_{k_3}(\omega_1 - \omega_2)}{(i(\omega_0 - \omega_2) + \gamma_a)} + c.c. \right]$$
Anticipating the future use of the rotation wave approximation applied to genuine normal modes of the system (see Eq. 3 below) I substituted \(2\omega_i (\omega - \omega_k + i\gamma_k)\) instead of \(\omega^2 - (\omega_k - i\gamma_k)^2\), neglected the non-resonant part of the linear susceptibility, and replaced all frequencies \(\omega\) in non-resonant expressions with atomic frequency \(\omega_0\). The latter approximation is justified because we will only consider the case where frequencies of all participating modes lie within a homogeneous line width of the lasing transition. Non-linear coupling parameters in Eq. 5, \(A_{kk_1,k_2k_3}\), are defined as

\[
A_{kk_1,k_2k_3} = \int \varepsilon(\mathbf{r}) f_k^* (\mathbf{r}) f_{k_1} (\mathbf{r}) f_{k_2} (\mathbf{r}) f_{k_3} (\mathbf{r}) d^3r
\]  

(6)

Lasing threshold and non-linear dynamics of the intensities

In order to illustrate the effects of spatial inhomogeneities on lasing threshold we find eigen frequencies of linearized Eq. (4) in a two-mode case. Imaginary parts of these frequencies, \(\Gamma_{1,2}\), are both positive below lasing threshold. With increasing pumping, however, one of them, \(\Gamma_1\), for instance, first changes its sign, and this point determines the lasing threshold. If we assume that \(\gamma_1 \ll \gamma_2\), a simple expression for the lasing threshold can be derived:

\[
\Delta N_{0r} = \frac{\Delta N_0}{V_{11}} \left[ 1 - \frac{V_{12}}{V_{11}} \right] ^2 \gamma_1 \gamma_2
\]  

(7)

where \(\Delta N_{0r}\) is a threshold value of \(\Delta N_0\) in a system with a uniform dielectric constant. Two effects of the non-uniformity appear in this expression. First, factor \(V_{11}\), which would be equal to unity for a uniform medium, affects the threshold even in the absence of the linear coupling between the modes. The value of this parameter depends on the spatial profile of the dielectric function; with an appropriate choice of the latter one can achieve a decrease in the lasing threshold. The second effect reflected in Eq. (7) is due to the coupling between the modes and results in further decrease of the threshold.

In order to derive rate equations we have to diagonalize the linear part of Eq. (4). To this end, we will, first, neglect the dispersion of the linear susceptibility. This approximation is justified if we are only interested in dynamics of intensities rather than lasing frequencies, and because all the frequencies of interest lie within the width of the atomic transition. After that we have to solve the eigen-value problem for the remaining matrix which is, however, essentially non-hermitian. Therefore, we have to find two adjoint sets of vectors — right \(\{c_i\}\) and left \(\{e_j\}\), which obey the bi-orthogonality condition \(\langle c_i | e_j \rangle = 0 \) when \(i \neq j\). In order to preserve standard expressions for intensities we shall normalize our right eigen-vectors using condition \(\langle e_i ^* | e_i \rangle = 1\) (so called power normalization\(11\)). As a result the product \(\alpha_i = \langle e_i | e_i \rangle \neq 1\). In order to eliminate linearly coupled terms from Eq. (4) we present cavity mode amplitudes, \(\tilde{E}_k\), as a linear combination of the right eigen vectors, \(\{ e_i \}\),

\[
\tilde{E}_k(\omega) = 2\pi \sum T_{ki} [Z_i(\omega)\delta(\omega - \Omega_i) + Z_i(\omega)\delta(\omega + \Omega_i)]
\]  

(8)

where columns of matrix \(T_{ki}\) are formed by the vectors \(\{ e_i \}\). In Eq. (5) we also introduced a slow changing amplitude approximation applied to the amplitudes of the true normal modes of the system. In the frequency domain, this approximation consists of presenting the amplitudes as a product of a frequency dependent part and a delta function, containing a lasing frequency \(\Omega_i\). Matrix \(T_{ki}\) is not a unitary matrix, and, transformation of Eq. (4) to the new basis has to rely on matrix \(\tilde{T}_{ki}\), whose rows consist of vectors \(\{ \tilde{e}_i \}\). The product of matrices \(T\) and \(\tilde{T}\) is a diagonal matrix with elements \(\alpha_i\). The resulting equations for slow amplitudes \(Z_i\) take the form

\[
-i \frac{dZ_i}{dt} + (\Omega_i - \Omega_j) Z_i = 2\pi \omega_0 \sum_j u_{ij} (\Pi_3) e^{-i(\Omega_j - \Omega_i)t}
\]  

(9)

where \(u_{ij} = \sum_{k,k_1} \tilde{T}_{ik} V_{kk_1} T_{k_1j} / \alpha_i\), and the non-linear contribution to polarization, in the new basis, is given by

\[
\Pi_3^{(3)} = i\mu |\Delta N_0 r|^2 \sum_{j,l,m} R_{ijklm} Z_j Z_l^* Z_m e^{-i(\Omega_j - \Omega_i - \Omega_m - \omega_0 + i\gamma_a)}
\]  

(10)

where we have neglected frequency dependence of the nonlinear coefficients \(R_{ijklm}\). Equations (9), (10), and (11) provide a basis for further analysis of the non-linear dynamics of the system under consideration.

In particular, the rate equations can be obtained in a standard way by separating real and imaginary parts of Eq. (11) and neglecting all oscillating terms on its right-hand side:

\[
\frac{dI_i}{dt} = 2I_i \left[ -\Gamma_i - \beta_i I_i - \sum_j \theta_{ij} I_j \right]
\]  

(12)

Here \(I_i\) is the dimensionless intensity of the \(i\)-th mode, \(\Gamma_i\) is its unsaturated amplification rate, \(\beta_i\) and \(\theta_{ij}\) are self- and cross-saturation parameters respectively, which are expressed in terms of coefficients \(R_{ijklm}\) of Eq. (11). These equations have the same form as standard lasing rate equations for a uniform medium, with the only difference being that instead of the combination of non-saturated gain and loss terms, we have a single parameter \(\Gamma_i\), representing the imaginary part of the mode’s eigen frequency. The main effect of the linear coupling is a renormalization of the non-linear coupling coefficients. The most
important feature of this renormalization, which makes it experimentally relevant, is a non-trivial dependence of the new coefficients on the intensity of pumping. This dependence arises because the coupling is carried by polarization, and, hence, its strength is proportional to the unsaturated inversion $\Delta N_0$. In order to illustrate the last point we consider an example of only two interacting modes. There are two possible regimes of behavior in this case: single mode lasing, when the mode competition prevents the second mode from lasing, and two-mode lasing, when the spatial hole burning prevails over the competition. The choice between these regimes is determined by a coupling parameter $C$, defined by the ratio of cross- and self-saturation parameters $C = (\beta_{12}\beta_{21})/(\beta_{11}\beta_{22})$. In a uniform medium, this parameter depends solely upon spatial distribution of the cavity modes, determined by the cavity's geometry. In the situation considered, here, this parameter becomes dependent on the pumping intensity. In order to illustrate the possible character of such dependence, we simulated parameter $C$ for a cavity which consists of two dielectric materials with different dielectric constants $\epsilon_1$ and $\epsilon_2$. The curve 1 on the figure shows the dependence $C(\Delta N_0)$ for two closest in frequency modes of such a cavity. The most striking feature of this graph is the steep decrease of this coefficient with $\Delta N_0$, which means that even if the modes of the empty cavity wouldn’t favor the spatial hole burning, the increasing with pumping linear interaction between the modes modifies their spatial structure in a way which is beneficial for the two-mode lasing. Similar behavior of $C(\Delta N_0)$ was also found for the dielectric constant of the shape $\epsilon(z) = \epsilon_0 + az^2$ or $\epsilon(z) = \epsilon_0 + \delta \cos z$, where $z$ is coordinate in the beam propagation direction. This effect might explain the two-mode behavior observed in random lasers. The fact that increased pumping can systematically drive $C$ below unity for various configurations of $\epsilon(z)$ makes such effect much more likely to occur in a random system than just a coincidental combination of various parameters suggested, for instance in Ref. [13].

A presence of the linear mode coupling in random lasers can be verified directly by observation of dependence of $I(\Delta N_0)$ in single and multi-mode regimes, which, if effects considered here are responsible for the observed multi-mode behavior, should deviate from a simple linear behavior expected in lasers with the uniform dielectric constant (see curve 2 in Fig.1).

**Conclusion** We derived non-linear equations describing dynamics of lasing modes in a cavity whose dielectric constant is spatially non-uniform in the direction of beam propagation. For a number of spatial profiles of $\epsilon(z)$ it is shown that the non-uniformity enhances spatial hole burning and promote two-mode lasing. This effect can explain recent observation of two-mode behavior in random lasers. The equations derived in the paper can also be used to manipulate properties of lasers through a design of spatial profile of the dielectric function.

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