Effects of Spatial Nonuniformity on Laser Dynamics

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Semiclassical equations of lasing dynamics are rederived for a lasing medium in a cavity with a spatially nonuniform dielectric constant. The nonuniformity 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 [1,2]. 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 [3] 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. [4] and consecutive works with strongly scattering systems (see recent reviews in Refs. [1,2]) led to an assumption that lasing observed in those experiment is due to the formation of prelocalized, if not completely localized, states of light, which play a role of lasing cavities [5] and provide *coherent resonant* optical feedback as opposed to *nonresonant feedback* affecting only intensity of light in the diffusion case. The presence of narrow multiple lasing peaks [4] as well as Poisson statistics of emitted radiation [6] were considered as evidence in the favor of this interpretation of these experiments. However, it was shown in Ref. [7] that the nonresonant feedback can also result in lasing with multiple narrow peaks. Moreover, the authors of Ref. [8] 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 Ref. [9] assume a particular significance. In these experiments a multipeak lasing was observed in poly(methyl methacrylate) (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, γ_a. This is naturally explained by the competition of modes "feeding" from the same population inversion and spectral hole burning in inhomogeneously broadened systems [9]. However, above certain value of the pumping intensity, there were observed two lasing peaks coexisting *within the homogeneous line width* γ_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 burning [10]. Such a behavior, however, cannot take place in the case of the nonresonant feedback, because in diffusive systems lasing only occurs at the frequency of an atomic transition [3] (multiple peaks in Ref. [7] are due to inhomogeneous broadening of the transition used to generate emission and do not signify a truly multimode behavior).

Thus, as of today, the results of Ref. [9] 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 nonlinear mode interaction in such systems. However, since the experiments of Ref. [9] deal with just a single realization, the randomness, by itself, is not important here. What is important is the spatial inhomogeneity of the quasicavity, supporting the modes of interest. The main objective of this Letter, 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 nonuniform cavities. Currently, there is a tremendous interest in lasing in systems with a modulated dielectric constant, for instance, photonic crystals. The results 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 Letter can be used to manipulate properties of lasers by using spatial dependence of the cavity dielectric function as a new design parameter.

A general multimode theory of lasing in systems with an arbitrarily inhomogeneous dielectric constant, $\epsilon(\mathbf{r})$, presented here is an extension of semiclassical Lamb theory [10] for the media whose dielectric constant is inhomogeneous *in the direction of propagation* of the laser beam (inhomogeneity in the perpendicular directions results in wave guiding effects, which are well studied in laser physics (see, for instance, [11]). This inhomogeneity modifies the orthonormalization condition for the eigenmodes of the cavity, making the standard inner product of the modes belonging to different eigenfrequencies different from zero. The main effect resulting from this nonorthogonality 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 nonorthogonality of eigenmodes due to the inhomogeneity of $\epsilon(\mathbf{r})$ should not be confused with nonorthogonality of Fox-Li modes of *uniform* but *leaky (open)* cavities, which arises due to *nonhermitian* nature of the respective eigenvalue problem and does not result in any additional linear coupling between the modes [12]. The only consequence of the nonhermitian 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 [13].

Multimode laser equations for an inhomogeneous medium.—We consider an ideal cavity specified by an inhomogeneous dielectric function $\epsilon(\mathbf{r})$ and some boundary conditions. The cavity is filled with an active medium characterized by its polarization $\mathbf{P}(\mathbf{r})$. Let us assume that we know the full system of eigenmodes, $f_k(\mathbf{r})$, and respective eigenfrequencies ω_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: $E = \Sigma_k E_k(t) f_k(\mathbf{r})$, $P = \Sigma_k P_k(t) f_k(\mathbf{r})$, 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 $\epsilon(\mathbf{r})$ [14]: $\int \epsilon(\mathbf{r}) f^*_{k_1}(\mathbf{r}) f_{k_2}(\mathbf{r}) d\mathbf{r} = \delta_{k_1 k_2}$, which means that the wave functions $f_k(\mathbf{r})$ themselves are neither normalized nor orthogonal. As a result, the dynamic equations for the amplitudes, E_k, takes the following form

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

where we introduced cavity losses, characterized by phenomenological parameters γ_k. The main peculiarity of Eq. (1) is the presence of the linear coupling between different polarization amplitudes P_k, characterized by nondiagonal elements of the matrix

$$V_{kk_1} = \int f^*_{k_1}(\mathbf{r}) f_{k_2}(\mathbf{r}) d\mathbf{r}. \quad (2)$$

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 equations can be, in principle, derived for inhomogeneous open cavities as well, where eigenmodes f_k should be replaced by appropriate Fox-Li modes. The hermitian orthogonality in this case is replaced by the biorthogonality, which involves adjoint set of modes. For a general case of nonuniform open cavity this condition was derived, for instance, in Ref. [15]. 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 γ_a^{-1}, and population relaxation time τ, and we use a standard density matrix approach in order to derive equations for polarization amplitudes P_k and population difference ΔN. 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 Ξ_k and Π_k are slowly changing amplitudes, and Ω_k is a frequency of the respective lasing mode. However, forcing this procedure onto Eq. (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 ΔN_0 represents nonsaturated 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 nonstationary 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:

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

where a tilde on top of a symbol signifies the Fourier transform of the respective quantity, and the polarization is separated into a linear and third-order nonlinear contribution, $\tilde{P}^{(3)}$. The former is taken into account in Eq. (4) by introducing a linear susceptibility $\chi^{(1)}(\omega)$, and the expression for the latter was derived in a standard way from the full system of density matrix equation using a standard perturbation approach.

$$\tilde{P}^{(3)}_k = \frac{|\mu|^4 \Delta N_0}{32\pi^2\hbar^3} \sum_{kk_1k_2k_3} A_{kk_1k_2k_3} \int d\omega_1 d\omega_2 \frac{\tilde{E}_{k_1}(\omega - \omega_1)}{(\omega - \omega_0 + i\gamma_a)(i\omega_1 - 1/\tau)} \left[\frac{\tilde{E}_{k_2}(\omega_2)\tilde{E}_{k_3}(\omega_1 - \omega_2)}{i(\omega_0 - \omega_2) + \gamma_a} + \text{c.c} \right]. \quad (5)$$

Anticipating the future use of the rotation wave approximation applied to genuine normal modes of the system (see Eq. (8) below) I substituted $2\omega(\omega - \omega_k + i\gamma_k)$ instead of $\omega^2 - (\omega_k - i\gamma_k)^2$, neglected the nonresonant part of the linear susceptibility, and replaced all frequencies ω in nonresonant expressions with atomic frequency ω_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. Nonlinear coupling parameters in Eq. (5), $A_{kk_1k_2k_3}$, are defined as

$$A_{kk_1k_2k_3} = \int \epsilon(\mathbf{r}) f_k^*(\mathbf{r}) f_{k_1}(\mathbf{r}) f_{k_2}(\mathbf{r}) f_{k_3}^*(\mathbf{r}) d^3 r. \qquad (6)$$

Lasing threshold and nonlinear dynamics of the intensities.—In order to illustrate the effects of spatial inhomogeneities on the lasing threshold we find eigenfrequencies of linearized Eq. (4) in a two-mode case. Imaginary parts of these frequencies, $\Gamma_{1,2}$ are both positive below the lasing threshold. With increasing pumping, however, one of them, Γ_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:

$$\Delta N_{0_{tr}} = \frac{\Delta N_{0_{tr}}^0}{V_{11}} \left[1 - \left(\frac{V_{12}}{V_{11}} \right)^2 \frac{\gamma_1}{\gamma_2} \right] \qquad (7)$$

where $N_{0_{tr}}^0$ is a threshold value of ΔN_0 in a system with a uniform dielectric constant. Two effects of the nonuniformity 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 eigenvalue problem for the remaining matrix, which is, however, essentially nonhermitian. Therefore, we have to find two adjoint sets of vectors—right ($|e_i\rangle$) and left ($\langle \tilde{e}_j|$), which obey the bi-orthogonality condition $\langle \tilde{e}_j | e_i \rangle = 0$ when $i \neq j$. In order to preserve standard expressions for intensities we shall normalize our right eigenvectors using condition $\langle e_i^* | e_i \rangle = 1$ (so-called power normalization [12]). As a result the product $\alpha_i = \langle \tilde{e}_i | e_i \rangle \neq 1$. 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 eigenvectors, $|e_i\rangle$,

$$\tilde{E}_k(\omega) = 2\pi \sum T_{ki}[Z_i(\omega)\delta(\omega - \Omega_i) + Z_i(\omega)\delta(\omega + \Omega_i)], \qquad (8)$$

where columns of matrix T_{ki} are formed by the vectors $|e_i\rangle$. In Eq. (8) 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 Ω_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 $\langle \tilde{e}_i|$. The product of matrices $\mathbf{T}$ and $\tilde{\mathbf{T}}$ is a diagonal matrix with elements α_i. The resulting equations for slow amplitudes Z_i take the form

$$-i\frac{dZ_i}{dt} + (\tilde{\Omega}_i - \Omega_i)Z_i = 2\pi\omega_0 \sum_j u_{ij} \Pi_j^{(3)} e^{-i(\Omega_j - \Omega_i)t}, \qquad (9)$$

where $u_{ij} = \Sigma_{k,k_1} \tilde{T}_{ik} V_{kk_1} T_{k_1 j}/\alpha_i$, and the nonlinear contribution to polarization, in the new basis, is given by

$$\Pi_i^{(3)} = \frac{|\mu|^4 \Delta N_0 \tau}{8\hbar^3 \alpha_i \gamma_a} \sum_{j,l,m} R_{ijlm} \frac{Z_j Z_l^* Z_m e^{-i(\Omega_j - \Omega_l + \Omega_m - \Omega_i)t}}{\Omega_j - \Omega_l + \Omega_m - \omega_0 + i\gamma_a}, \qquad (10)$$

$$R_{ijlm} = -\sum_{k_i} \tilde{T}_{ik} A_{kk_1k_2k_3} T_{k_1 m}[T_{k_2 j}(T_{k_3 l})^* + T_{k_3 j}(T_{k_2 l})^*] \qquad (11)$$

where we have neglected frequency dependence of the nonlinear coefficients R_{ijlm}. Equations (9)–(11) provide a basis for further analysis of the nonlinear 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. (9) and neglecting all oscillating terms on its right-hand side:

$$\frac{dI_i}{dt} = 2I_i[-\Gamma_i - \beta_i I_i - \sum_j \theta_{ij} I_j] \qquad (12)$$

Here I_i is the dimensionless intensity of the ith mode, Γ_i is its unsaturated amplification rate, and β_i and θ_{ij} are self- and cross-saturation parameters, respectively, which are expressed in terms of coefficients R_{ijlm} 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 nonsaturated gain and loss terms, we have a single parameter Γ_i, representing the imaginary part of the mode's eigenfrequency. The main effect of the linear coupling is a renormalization of the nonlinear coupling coefficients. The most important feature of this renormalization, which makes it experimentally relevant, is a nontrivial dependence of the new coef-

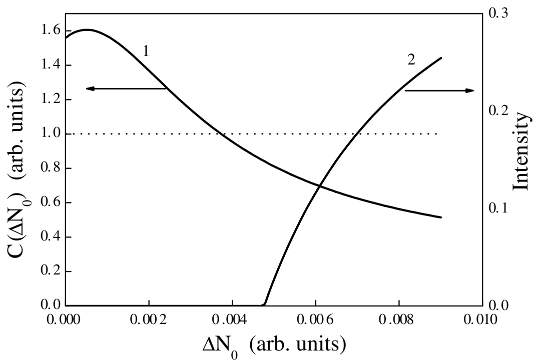

FIG. 1. Dependence of the nonlinear coupling parameter C on ΔN_0 (curve 1) and deviation of the intensity in a single mode regime from a linear dependence on pumping (curve 2).

ficients 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 ΔN_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 $C = (\theta_{12}\theta_{21})/(\beta_{11}\beta_{22})$ [10]. 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 ϵ_1, and ϵ_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 ΔN_0, which means that even if the modes of the empty cavity would not 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 [9]. The fact that increased pumping can systematically drive C below unity for various configurations of $\epsilon(\mathbf{r})$ makes such effects much more likely to occur in a random system than just a coincidental combination of various parameters suggested, for instance in Ref. [16]. 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 multimode regimes, which, if effects considered here are responsible for the observed multimode 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 nonlinear equations describing dynamics of lasing modes in a cavity whose dielectric constant is spatially nonuniform in the direction of beam propagation. For a number of spatial profiles of $\epsilon(\mathbf{r})$ it is shown that the nonuniformity enhances spatial hole burning and promotes 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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