Perturbation approach in Heisenberg equations for lasers

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Nonlinear Heisenberg-Langevin equations are solved analytically by operator Fourier expansion for the laser in the light-emitting diode (LED) regime. Fluctuations of populations of lasing levels are taken into account as perturbations. The spectra of operator products are calculated as convolutions, preserving Bose commutations for the lasing field operators. It is found that fluctuations of population significantly affect spontaneous and stimulated emissions into the lasing mode, increase the radiation rate, the number of lasing photons, and broaden the spectrum of a bad cavity threshold-less and the superradiant lasers. The method can be applied to various resonant systems in quantum optics.

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I. INTRODUCTION

Operator Heisenberg-Langevin equations (HLE), as quantum Maxwell-Bloch equations, are widely used in quantum optics and laser physics [1]. They are applied for modeling devices and processes in nonlinear optic [2,3], lasers [4–7], the generation of nonclassical light [8], qubits [9], and other quantum phenomena [10], making them an important part of physics [11]. HLEs are in the background of various theoretical methods of quantum optics such as the input-output theory [12,13] and the cluster expansion method [14,15].

HLE for lasers and resonant optical systems are often nonlinear in operators, which makes them difficult to solve analytically. This paper continues and extends the research of the authors of [16] on analytically solving HLEs for lasers.

Several methods of solving HLE were proposed [17–23]. A relatively simple and widespread method of solving HLE in quantum optics and laser physics [4–7,24,25] is a generalization of the perturbation approach of the classical oscillation theory [26]. This is the linearization of HLE around the mean values of operators and solving linear equations for operators of small perturbations.

Consider, for example, the nonlinear term $\hat{a}\hat{N}_e$ in Eq. (4b) of the laser model in Sec. II, where $\hat{a}$ is a Bose operator of the lasing field amplitude and $\hat{N}_e$ is the operator of the population of excited states of lasing transitions. $\hat{N}_e$ can be separated on the mean N_e and fluctuations $\delta\hat{N}_e$: $\hat{N}_e = N_e + \delta\hat{N}_e$. Supposing that the contribution of fluctuations $\delta\hat{N}_e$ is small and can be neglected, we approximately replace $\hat{a}\hat{N}_e$ by the term $\hat{a}N_e$ linear in the operator $\hat{a}$. Then the stationary HLE for the laser in Sec. II is linearized and can be solved as in [7,16,27] at a weak excitation of the laser, when the laser does not generate coherent radiation, and the mean amplitude of the lasing field $a = 0$. This approach reproduces well-known results, such as the laser linewidth [16], and leads to new results, such as the collective Rabi splitting [27], but it must be extended for considering population fluctuations at the weak excitation of the laser.

In a similar way the laser HLE can be linearized and solved for a high excitation, when the laser does generate coherent radiation, so $\hat{a} \approx a + \delta\hat{a}$, where $\delta\hat{a}$ is the operator of a small perturbation [5,6,16]. In this case, population fluctuations are taken into account and lead to well-known relaxation oscillations' peaks in the intensity fluctuation spectra [25] and to the prediction of such peaks in the field spectra of the bad cavity nanolasers [16].

A direct generalization of the standard perturbation approach for considering population fluctuations at a low excitation meets difficulties. Consider, for example, the laser at a weak excitation, when the mean laser field $a = 0$. Following the standard procedure of the classical perturbation theory we neglect $\delta\hat{N}_e$ and find a zero-order solution $\hat{a} = \hat{a}_0$ [7,16,27]. Next we must replace $\hat{a}\delta\hat{N}_e$ with the linear term $\hat{a}_0\delta\hat{N}_e$ and obtain linear equations with the time-dependent operator coefficients, like $\hat{a}_0$. It is unclear how to solve such equations.

To overcome such a difficulty, in [16] we replaced $\hat{a}_0$ in $\hat{a}_0\delta\hat{N}_e$ by $\sqrt{n}$, where n is the mean photon number. This approach made a "smooth transition" between the high and the low excitations of the laser, but remained without a justification for the low excitation in [16]. It was mentioned in [16] that the approach is good if the population fluctuations with the low excitation are negligibly small (we will see that this is not always the case). The features of lasing, found in [16] due to the population fluctuations at the low excitation, need to be provem with a more rigorous approach.

One purpose of this work is to extend the analysis of [16] and consider population fluctuations rigorously at the low excitation, when the laser works in the LED regime. We will correct some results of [16] related with the population fluctuations in the LED regime.

As we outlined above, it is difficult to take into account population fluctuations in the nonlinear laser HLEs at the low excitation with the standard perturbation approach. Another purpose of the paper is to formulate a perturbation approach

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for solving nonlinear stationary HLEs at the low excitation of the laser in the first order on population fluctuations.

Only a few methods can be applied in the higher order on quantum perturbations as, for example, a cluster expansion method [14,15]. It allows for finding mean values of high-order correlations of products of operators, but it does not calculate the spectra of optical fields. Path integral formalism can be used in some problems of nonlinear and quantum optics [28,29]. However, it is applied mostly to systems with quadratic Hamiltonian, i.e., to linear systems. Quantum perturbation theory, in time, is often applied for the analysis of nonstationary processes in nonlinear optics [30], and it is restricted by short periods of time when the effect of nonlinear terms is negligibly small.

Here we consider the population fluctuations as a perturbation using the operator Fourier expansion, and express the power spectra of the operator products as convolutions of spectra of multipliers in the product.

An important part of the method is preserving commutation relations for Bose operators of the field. This lets us to take into account quantum fluctuations in the field with a small number of photons.

Because of the dissipation and fluctuations, the oscillation spectra of resonant systems are bands centered at mode frequencies. We suppose, as usual, that the width of the band is much smaller than the mode frequency and use a rotating wave approximation (RWA) [31].

As usual, we suppose that the laser interacts with incoherent "white noise" baths of broad spectra.

We demonstrate the method in the example of the quantum model of a single-mode laser with a homogeneously broadened active medium of two-level emitters, the same as in [16,27]. We suppose a large number of emitters $N_0 \gg 1$ and consider the LED radiation regime at a weak excitation of the laser, when the mean number n of lasing photons is small $n < 1$ or of the order of 1, so the laser does not generate coherent radiation.

We will show that population fluctuations increase, at certain conditions, the radiation rate into the lasing mode; increasing the number of lasing photons and broad lasing spectra. This can be seen most clearly in lasers with low-quality cavities and large gain where population fluctuations are high and collective effects, as a superradiance, are important [32–34]. Such superradiant lasers were experimentally realized, for example, with cold alkaline earth atoms [35–38], rubidium atoms [39], and with quantum dots [14].

Quantum models of a laser were presented in many papers and books as, for example, [4,40,41]. Among popular methods of the laser theory are the linearization of Heisenberg-Langevin equations around the steady state [5,6,40], solving the master equation for the density matrix [4] or Lindblad master equation [42]. The method proposed here has not been used before.

Usual perturbation theory with the linearization of operator equations on small fluctuations around the steady states is widely used in the laser quantum rate equation theory [25,43–46]. Quantum rate equations for lasers are valid with the adiabatic elimination of the polarization of the lasing media. The method presented here does not require the adiabatic elimination of polarization, so it can be applied for the modeling of lasers with bad cavities and collective effects.

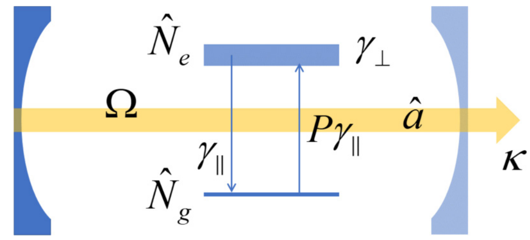

FIG. 1. Scheme of the two-level laser. Upper levels of emitters with the population operator $\hat{N}_e$ decay to the low levels with the rate $\gamma_\parallel$ and pumped with the rate $\gamma_\parallel P$ from the low levels with the population $\hat{N}_g$. The width of the lasing transition is $\gamma_\perp$. Lasing mode described by Bose-operator $\hat{a}$ decays through the semitransparent mirror with the rate κ and resonantly interacts with lasing transitions of two-level emitters with the vacuum Rabi frequency Ω.

In this paper we do not provide rigorous mathematical justification of the method, in particular, we do not prove its conversion to the exact solution. Our aim is to demonstrate basic physical ideas and to show the application of the method. We will use general properties of Heisenberg representation and well-known results of quantum mechanics [47] for the derivation of the mathematical part of the method in Appendixes A and B.

We demonstrate the method on the example of the laser model described in Sec. II. There we derive the laser HLE and obtain from them equations for Fourier-component operators.

In Sec. III we apply the perturbation approach to the laser model in the zero-order approximation, when population fluctuations are neglected. In Sec. IV we solve the laser equations, taking into account population fluctuations in the first-order approximation. We demonstrate the important parts of the method: the calculation of the spectrum of the operator product with convolutions and preserving Bose-commutation relations for the lasing field operator. Section V presents and discuses results related with the effect of population fluctuations on the lasing in the LED regime at low excitation. We show that population fluctuations increase the spontaneous and the stimulated emission rates into the lasing mode, leading to the increase of the number of lasing photons; they broad the lasing field spectra, but do not lead to narrow peaks in the field spectra found in [16]. Such peaks are the consequence of the application of the standard perturbation approach at the low excitation. Results are summarized in Sec. VI. Appendix A shows the Fourier expansion for operators, Appendix B calculates the spectrum of the operator product, and Appendix C calculates diffusion coefficients. Appendix D presents equations for population fluctuations for the calculation of the population fluctuation spectrum and the justification of the approximation (37).

II. EQUATIONS FOR TWO-LEVEL LASER

We consider a quantum model of a single mode homogeneously broadened laser in the stationary regime with $N_0 \gg 1$ two-level identical emitters, the same as in [16,27], shown schematically in Fig. 1. Lasing transitions are in the exact resonance with the cavity mode with the optical frequency ω_0. $\hat{a}(t)e^{-i\omega_0 t}$ is the Bose operator of the lasing mode and the operator $\hat{a}(t)$ of the complex amplitude is changed much more slowly than $e^{-i\omega_0 t}$.

The Hamiltonian of the laser, written in the interaction picture with the carrier frequency ω_0 and in the RWA approximation, is

$$H = i\hbar\Omega \sum_{i=1}^{N_0} f_i(\hat{a}^\dagger\hat{\sigma}_i - \hat{\sigma}_i^\dagger\hat{a}) + \hat{\Gamma}. \tag{1}$$

Here Ω is the vacuum Rabi frequency, f_i describes the difference in couplings of different emitters with the lasing mode. $\hat{\sigma}_i$ is a lowing operator of ith emitter, $\hat{\Gamma}$ describes the interaction of the mode and emitters with the white noise baths of the environment.

The commutation relations for the operators are

$$[\hat{a}, \hat{a}^\dagger] = 1, \quad [\hat{\sigma}_i, \hat{\sigma}_j^\dagger] = \left(\hat{n}_i^g - \hat{n}_i^e\right)\delta_{ij},$$
$$\left[\hat{\sigma}_i, \hat{n}_j^e\right] = \left[\hat{n}_j^g, \hat{\sigma}_i\right] = \delta_{ij}\hat{\sigma}_i, \tag{2}$$

where $\hat{n}_j^e$ and $\hat{n}_j^g$ are operators of populations of the upper and the low levels of the ith emitter and δ_{ij} is the Kronecker symbol.

We introduce operators $\hat{v}$ and $\hat{N}_{e,g}$ of the polarization and populations of all emitters

$$\hat{v} = \sum_{i=1}^{N_0} f_i\hat{\sigma}_i, \quad \hat{N}_{e,g} = \sum_{i=1}^{N_0} \hat{n}_i^{e,g}. \tag{3}$$

Using commutation relations (2) and Hamiltonian (1) we write Maxwell-Bloch equations for $\hat{a}$, $\hat{v}$, and $\hat{N}_e$

$$\dot{\hat{a}} = -\kappa\hat{a} + \Omega\hat{v} + \hat{F}_a, \tag{4a}$$
$$\dot{\hat{v}} = -(\gamma_\perp/2)\hat{v} + \Omega f\hat{a}(2\hat{N}_e - N_0) + \hat{F}_v, \tag{4b}$$
$$\dot{\hat{N}}_e = -\Omega\hat{\Sigma} + \gamma_\parallel[P(N_0 - \hat{N}_e) - \hat{N}_e] + \hat{F}_{N_e}, \tag{4c}$$

where

$$\hat{\Sigma} = \hat{a}^\dagger\hat{v} + \hat{v}^\dagger\hat{a}, \tag{5}$$

κ, $\gamma_\perp$, and $\gamma_\parallel$ are decay rates; $P\gamma_\parallel$ is the pump rate; and $\hat{F}_\alpha$ with the index $\alpha = \{a, v, N_e\}$ are Langevin forces. The total number of emitters is preserved, so $\hat{N}_e + \hat{N}_g = N_0$.

In Eqs. (4) and below we approximate $f_i^2 \approx f = N_0^{-1}\sum_{i=1}^{N_0} f_i^2$ and use notations with a "hat" for operators and without a hat for mean values as, for example, $N_e = \langle\hat{N}_e\rangle$.

We separate the mean values and fluctuations in population operators $\hat{N}_{e,g} = N_{e,g} + \delta\hat{N}_{e,g}$, in $\hat{\Sigma} = \Sigma + \delta\hat{\Sigma}$, insert them into Eqs. (4) and write

$$\dot{\hat{a}} = -\kappa\hat{a} + \Omega\hat{v} + \hat{F}_a, \tag{6a}$$
$$\dot{\hat{v}} = -(\gamma_\perp/2)\hat{v} + \Omega f(\hat{a}N + 2\hat{a}\delta\hat{N}_e) + \hat{F}_v, \tag{6b}$$
$$\delta\dot{\hat{N}}_e = -\Omega\delta\hat{\Sigma} - \gamma_P\delta\hat{N}_e + \hat{F}_{N_e}, \tag{6c}$$

where $\gamma_P = \gamma_\parallel(P+1)$. With the derivation of Eqs. (6c) we take

$$0 = -\Omega\Sigma + \gamma_\parallel[P(N_0 - N_e) - N_e]. \tag{7}$$

In Eq. (6b) and below $N = N_e - N_g$ is the mean population inversion.

We take the stationary mean photon number $n = \langle\hat{a}^\dagger\hat{a}\rangle$ and find from Eq. (6a)

$$0 = -2\kappa n + \Omega\Sigma. \tag{8}$$

Equations (8) and (7) lead to the energy conservation law

$$2\kappa n = \gamma_\parallel[P(N_0 - N_e) - N_e]. \tag{9}$$

In the next sections we consider population fluctuations $\delta\hat{N}_e$ as a perturbation and solve the stationary Eqs. (6) approximately using Fourier expansion for the operators

$$\hat{\alpha}(t) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}\hat{\alpha}(\omega)e^{-i\omega t}d\omega, \tag{10}$$

where $\hat{\alpha}$ denotes an operator $\hat{\alpha} = \hat{a}, \hat{v}, \ldots$. In particular, $\hat{\alpha}$ can be the product of operators $\hat{a}\delta\hat{N}_e$. $\hat{\alpha}(\omega)$ is the Fourier component of the operator $\hat{\alpha}(t)$. $\hat{\alpha}(\omega)$ can be expressed through $\hat{\alpha}(t)$ by the reverse Fourier transform; see more about the operator Fourier expansion in Appendix A.

In the stationary case

$$\langle\hat{\alpha}^\dagger(\omega)\hat{\alpha}(\omega')\rangle = S_{\alpha^\dagger\alpha}(\omega)\delta(\omega + \omega'), \tag{11}$$

where $S_{\alpha^\dagger\alpha}(\omega)$ is a power spectrum of fluctuations, corresponding to $\hat{\alpha}(t)$. We will find power spectra solving equations for Fourier-component operators and using relations as in Eq. (11). A similar way of calculations of field spectra can be found in the literature, for example, in [44,48,49]. It can be shown that $S_{\alpha^\dagger\alpha}(\omega)$ in Eq. (11) is a Fourier component of the autocorrelation function $\langle\hat{\alpha}^\dagger(t+\tau)\hat{\alpha}(t)\rangle$ in accordance with the Wiener-Khinchin theorem [50,51].

Fourier expansion for operators is widely used in laser physics and quantum optics [5–7,13,44,45,48], as well as in the classical stochastic theory [52]. However, the Fourier expansion of a stochastic function is not well defined [50,51], so quite often the calculation of the power spectra, as $S_{\alpha^\dagger\alpha}(\omega)$, is carried out without the use of Fourier-component operators. Instead, one calculates a time-dependent autocorrelation function and then applies the Wiener-Khinchin theorem [24,53–55]. In our opinion, the calculation of spectra in the stationary case with Fourier-component operators and the formula (11) (see examples in [25,45,48]) is more easy than with the Wiener-Khinchin theorem. However, the operator Fourier expansion (10) must be justified, so in Appendix A we make the operator Fourier expansion (10) based on quantum-mechanical relations in the Heisenberg picture in the stationary case.

Making the Fourier expansion (10) in Eqs. (6) we obtain algebraic equations for Fourier-component operators

$$0 = (i\omega - \kappa)\hat{a}(\omega) + \Omega\hat{v}(\omega) + \hat{F}_a(\omega), \tag{12a}$$
$$0 = (i\omega - \gamma_\perp/2)\hat{v}(\omega) + \Omega f[\hat{a}(\omega)N + 2(\hat{a}\delta\hat{N}_e)_\omega] + \hat{F}_v(\omega), \tag{12b}$$
$$0 = (i\omega - \gamma_P)\delta\hat{N}_e(\omega) - \Omega\delta\hat{\Sigma}(\omega) + \hat{F}_{N_e}(\omega). \tag{12c}$$

Here $(\hat{a}\delta\hat{N}_e)_\omega$ is a Fourier component of the operator product $\hat{a}(t)\delta\hat{N}_e(t)$.

Correlations for the Fourier components of Langevin forces $\hat{F}_\alpha(\omega)$, $\hat{F}_\beta(\omega)$ are

$$\langle \hat{F}_\alpha(\omega)\hat{F}_\beta(\omega')\rangle = 2D_{\alpha\beta}\delta(\omega+\omega'), \tag{13}$$

where $2D_{\alpha\beta}$ is a spectral power density of the bath noise or a diffusion coefficient. The diffusion coefficients

$$2D_{aa^\dagger} = 2\kappa, \quad 2D_{a^\dagger a} = 0, \tag{14}$$

correspond to the lasing mode-harmonic oscillator [12,13]. They remain the same in any order of our approach. We choose diffusion coefficients $2D^{(i)}_{v^\dagger v}$ and $2D^{(i)}_{vv^\dagger}$ such that Bose-commutation relations for the operator $\hat{a}$ of the lasing mode will be preserved in the $i = 0, 1, \dots,$ order of the approximation on population fluctuations.

III. ZERO-ORDER APPROXIMATION

In the zero-order approximation we neglect population fluctuations [7,16,27]. We drop the term $(\hat{a}\delta\hat{N}_e)_\omega$ in Eq. (12b) and take the Langevin force $\hat{F}_v(\omega) = \hat{F}^{(0)}_v(\omega)$ with diffusion coefficients

$$2D^{(0)}_{v^\dagger v} = f\gamma_\perp N_e, \quad 2D^{(0)}_{vv^\dagger} = f\gamma_\perp N_g. \tag{15}$$

These diffusion coefficients are found at the absence of population fluctuations in Appendix C.

In the zero-order approximation $\hat{a} = \hat{a}_0$. We solve the set of Eqs. (12a) and (12b), taken without $(\hat{a}\delta\hat{N}_e)_\omega$, and find

$$\hat{a}_0(\omega) = \frac{(\gamma_\perp/2 - i\omega)\hat{F}_a(\omega) + \Omega\hat{F}_v(\omega)}{s(\omega)}, \tag{16}$$

where

$$s(\omega) = (i\omega-\kappa)(i\omega-\gamma_\perp/2) - (\kappa\gamma_\perp/2)N/N_{\rm th}, \tag{17}$$

and $N_{\rm th} = \kappa\gamma_\perp/2\Omega^2 f$ is a threshold population inversion found in the semiclassical laser theory [16,27].

The spectrum $n_0(\omega)$ of the lasing field satisfies

$$\langle \hat{a}^\dagger_0(\omega)\hat{a}_0(\omega')\rangle = n_0(\omega)\delta(\omega+\omega'). \tag{18}$$

We calculate $n_0(\omega)$ from Eqs. (16) and (18) and using diffusion coefficients (14) and (15)

$$n_0(\omega) = \frac{(\kappa\gamma_\perp^2/2)N_e/N_{\rm th}}{S(\omega)}, \tag{19}$$

where $S(\omega) = |s(\omega)|^2$. The mean photon number $n_0 = (2\pi)^{-1}\int_{-\infty}^{\infty} n_0(\omega)d\omega$ is

$$n_0 = \frac{N_e}{(1+2\kappa/\gamma_\perp)(N_{\rm th}-N)}. \tag{20}$$

To ensure that Bose-commutation relations $\langle[\hat{a}_0,\hat{a}^\dagger_0]\rangle = 1$ are satisfied, we find the spectrum $(n_0+1)_\omega$ such that $\langle \hat{a}_0(\omega)\hat{a}^\dagger_0(\omega')\rangle = (n_0+1)_\omega\delta(\omega+\omega')$

$$(n_0+1)_\omega = \frac{2\kappa(\omega^2+\gamma_\perp^2/4) + (\kappa\gamma_\perp^2/2)N_g/N_{\rm th}}{S(\omega)}, \tag{21}$$

and the spectrum of the commutator $\langle[\hat{a}_0(\omega),\hat{a}^\dagger_0(\omega')]\rangle = [\hat{a}_0,\hat{a}^\dagger_0]_\omega\delta(\omega+\omega')$

$$[\hat{a}_0,\hat{a}^\dagger_0]_\omega = (n_0+1)_\omega - n_0(\omega). \tag{22}$$

The calculation shows that $(2\pi)^{-1}\int_{-\infty}^{\infty}[\hat{a}_0,\hat{a}^\dagger_0]_\omega d\omega = 1$, so the Bose-commutation relations for $\hat{a}_0$ are satisfied.

IV. FIRST-ORDER APPROXIMATION

In the first-order approximation we denote $\hat{a} = \hat{a}_1$, keeping in Eq. (12b) the term $(\hat{a}_0\delta\hat{N}_e)_\omega$ with $\hat{a}$ replaced by $\hat{a}_0$ and take Langevin force $\hat{F}_v(\omega) = \hat{F}^{(1)}_v(\omega)$ with diffusion coefficients

$$2D^{(1)}_{v^\dagger v} = f\gamma_\perp[N_e + N_1(\omega)],$$
$$2D^{(1)}_{vv^\dagger} = f\gamma_\perp[N_g - N_1(\omega)]. \tag{23}$$

$N_1(\omega)$ in Eqs. (23) is added for satisfying the Bose-commutation relations $\langle[\hat{a}_1,\hat{a}^\dagger_1]\rangle = 1$. Expressions (23) are written such that the sum $2D^{(1)}_{v^\dagger v} + 2D^{(1)}_{vv^\dagger}$ does not depend on $N_1(\omega)$ and, therefore, on population fluctuations, as it is shown in Appendix C. This is why the same N_1 appears in both diffusion coefficients $2D^{(1)}_{v^\dagger v}$ and $2D^{(1)}_{vv^\dagger}$.

Solving the set of Eqs. (12a) and (12b) with $(\hat{a}_0\delta\hat{N}_e)_\omega$ and $\hat{F}^{(1)}_v(\omega)$ instead of $(\hat{a}\delta\hat{N}_e)_\omega$ and $\hat{F}_v(\omega)$, respectively, we find the Fourier-component operator

$$\hat{a}_1(\omega) = \hat{a}^{(1)}_0 + \frac{\kappa\gamma_\perp}{N_{th}}\frac{(\hat{a}_0\delta\hat{N}_e)_\omega}{s(\omega)}. \tag{24}$$

where $\hat{a}^{(1)}_0(\omega)$ is given by Eq. (16) with $\hat{F}_v(\omega) = \hat{F}^{(1)}_v(\omega)$.

Now we find $(\hat{a}_0\delta\hat{N}_e)_\omega$ and $N_1(\omega)$. We consider the spectrum $S_{a_0N_e}(\omega)$ of the operator product $\hat{a}_0\delta\hat{N}_e$

$$\langle(\hat{a}^\dagger_0\delta\hat{N}_e)_\omega(\hat{a}_0\delta\hat{N}_e)_{\omega'}\rangle = S_{a_0N_e}(\omega)\delta(\omega+\omega'). \tag{25}$$

We calculate $S_{a_0N_e}(\omega)$ neglecting cumulants in correlations, as in a well-known cumulant-neglect closure method in the classical statistical theory [56,57] and in the quantum cluster-expansion method [15]. In these methods the mean of, for example, four-operator products is approximated by the sum of products of the nonzero two-operator means. In the case of Eq. (25) this is

$$\langle \hat{a}^\dagger_0(\omega_1)\delta\hat{N}_e(\omega_2)\hat{a}_0(\omega_3)\delta\hat{N}_e(\omega_4)\rangle$$
$$\approx \langle \hat{a}^\dagger_0(\omega_1)\hat{a}_0(\omega_3)\rangle\langle\delta\hat{N}_e(\omega_2)\delta\hat{N}_e(\omega_4)\rangle, \tag{26}$$

since $\langle \hat{a}^\dagger_0(\omega_1)\delta\hat{N}_e(\omega_2)\rangle = 0$ and $\langle \hat{a}_0(\omega_1)\delta\hat{N}_e(\omega_2)\rangle = 0$ at the low excitation of the laser.

It is shown in Appendix B that $S_{a_0N_e}(\omega)$ calculated with the approximation (26) is a convolution $S_{a_0N_e}(\omega) = (n_0 * \delta^2N_e)_\omega$,

$$(n_0 * \delta^2N_e)_\omega = \frac{1}{2\pi}\int_{-\infty}^{\infty} n_0(\omega-\omega')\delta^2N_e(\omega')d\omega', \tag{27}$$

where $\delta^2N_e(\omega)$ is a spectrum of population fluctuations

$$\langle\delta\hat{N}_e(\omega)\delta\hat{N}_e(\omega')\rangle = \delta^2N_e(\omega)\delta(\omega+\omega'). \tag{28}$$

The field spectrum $n_1(\omega)$, $\langle\hat{a}^\dagger_1(\omega)\hat{a}_1(\omega')\rangle = n_1(\omega)\delta(\omega+\omega')$, can be represented, with the help of Eq. (24), as

$$n_1(\omega) = n_0(\omega) + n_{\rm sp}(\omega) + n_{st}(\omega). \tag{29}$$

Here $n_0(\omega)$, given by Eq. (19), is caused by the vacuum fluctuations of the lasing mode and the active medium polarization;

$$n_{\rm sp}(\omega) = \frac{\kappa\gamma_\perp^2}{2N_{\rm th}}\frac{N_1(\omega)}{S(\omega)} \tag{30}$$

is due to the effect of the population fluctuations on spontaneous emission: we see that $n_{\rm sp}(\omega)$ does not depend explicitly on the mean photon number;

$$n_{\rm st}(\omega) = \left(\frac{\kappa\gamma_\perp}{N_{\rm th}}\right)^2 \frac{(n_0 * \delta^2 N_e)_\omega}{S(\omega)} \tag{31}$$

is proportional to the mean photon number n_0, appeared in $(n_0 * \delta^2 N_e)_\omega$ and, therefore, it is due to the effect of the population fluctuations on the stimulated emission.

Replacing $(n_0 * \delta^2 N_e)_\omega$ by $n_0\delta^2 N_e(\omega)$ in Eq. (31) we come to the approach of [16], which is good if the field spectrum $n_0(\omega)$ is much narrower than the population fluctuation spectrum $\delta^2 N_e(\omega)$. This is true for the high excitation, when the laser generates coherent radiation, so $n_0(\omega) \approx n_0\delta(\omega)$ where $\delta(\omega)$ is the Dirac delta function. The term $n_{\rm sp}(\omega)$ does not appear in the approach of the authors of [16], which does not take into account the influence of population fluctuations on the spontaneous emission into the lasing mode.

With the derivation of Eqs. (29) to (31) we suppose that $\hat{a}_0\delta\hat{N}_e$, in the first-order approximation, is not correlated with $\hat{F}_a$ and $\hat{F}_v^{(1)}$.

We find $N_1(\omega)$ demanding Bose commutation relations $\langle[\hat{a}_1, \hat{a}_1^\dagger]\rangle = 1$. From Eq. (24) we obtain

$$[\hat{a}_1, \hat{a}_1^\dagger]_\omega = [\hat{a}_0, \hat{a}_0^\dagger]_\omega + \{(\kappa\gamma_\perp/N_{\rm th})^2([\hat{a}_0, \hat{a}_0^\dagger] * \delta^2 N_e)_\omega - \kappa\gamma_\perp^2 N_1(\omega)/N_{th}\}/S(\omega), \tag{32}$$

with the spectrum $[\hat{a}_0, \hat{a}_0^\dagger]_\omega$ given by Eq. (22). We know that $(2\pi)^{-1}\int_{-\infty}^{\infty}[\hat{a}_0, \hat{a}_0^\dagger]_\omega d\omega = 1$. Therefore $(2\pi)^{-1}\int_{-\infty}^{\infty}[\hat{a}_1, \hat{a}_1^\dagger]_\omega d\omega = 1$, if the nominator in the second term on the right in Eq. (32) is zero, which is true when

$$N_1(\omega) = (\kappa/N_{\rm th})([\hat{a}_0, \hat{a}_0^\dagger] * \delta^2 N_e)_\omega. \tag{33}$$

Inserting $N_1(\omega)$ from Eq. (33) into Eq. (30) we find

$$n_{\rm sp}(\omega) = \left(\frac{\kappa\gamma_\perp}{N_{\rm th}}\right)^2 \frac{([\hat{a}_0, \hat{a}_0^\dagger]/2 * \delta^2 N_e)_\omega}{S(\omega)}. \tag{34}$$

We see that $n_{\rm sp}(\omega)$ depends on the convolution of the population fluctuation spectrum $\delta^2 N_e(\omega)$ with the spontaneous emission noise spectrum. Indeed, the spectrum $[\hat{a}_0, \hat{a}_0^\dagger]_\omega/2$, in the convolution in Eq. (34), is a spectrum of vacuum field fluctuations in the lasing mode, or a "spectrum of the half of a photon": $(2\pi)^{-1}\int_{-\infty}^{\infty}([\hat{a}_0, \hat{a}_0^\dagger]_\omega/2)d\omega = 1/2$.

To find $n_{\rm sp}(\omega)$ and $n_{\rm st}(\omega)$ we must know the spectrum of the population fluctuations $\delta^2 N_e(\omega)$. From Eq. (12c) we find $\delta\hat{N}_e(\omega)$ and the population fluctuation spectrum

$$\delta^2 N_e(\omega) = \frac{\Omega^2\delta^2\Sigma(\omega) + 2D_{N_eN_e}}{\omega^2 + \gamma_P^2}, \tag{35}$$

where $\delta^2\Sigma(\omega)$ is the spectrum of $\delta\hat{\Sigma}(\omega)$. With calculations of $\delta^2 N_e(\omega)$ we use the same approximation as in [16], neglecting by correlations between the polarization and population fluctuations, i.e., between $\hat{F}_v$ and $\hat{F}_{N_e}$, which is a good approximation at a large number of emitters $N_0 \gg 1$. The diffusion coefficient $2D_{N_eN_e} = \gamma_\parallel(PN_g + N_e)$ is the same as in the rate equation laser theory [43].

We find $\delta\hat{\Sigma}(\omega)$ from Eqs. (D5) written Appendix D in the zero-order approximation on $\delta\hat{N}_e$. Then we find the spectrum $\delta^2\Sigma(\omega)$ from Eq. (D6). The explicit expression for $\delta^2\Sigma(\omega)$ is cumbersome so we do not present it here. With $\delta^2\Sigma(\omega)$ we integrate the spectrum (35) over frequencies and find the population fluctuation dispersion $\delta^2 N_e$.

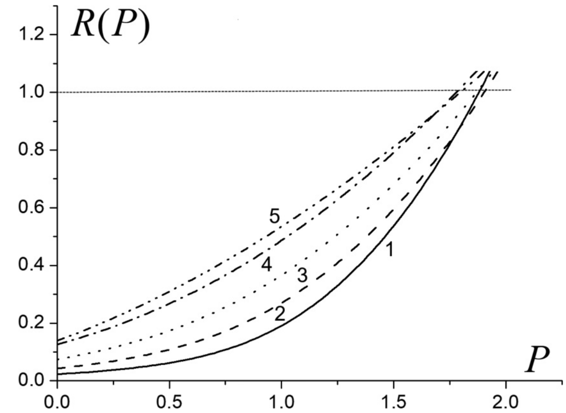

FIG. 2. The relative difference $R(P)$ of the population fluctuation dispersion found with and without $\delta\hat{\Sigma}$ for $\gamma_\perp = 5$ (curve 1), 10 (2), 20 (3), 50 (4), and 500 (5). $R(P) < 1$, so population fluctuations caused by $\delta\hat{\Sigma}$ [the first term in Eq. (35)] is smaller than population fluctuations caused by the second term in Eq. (35) at the weak excitation, when the pump rate $P < 2$.

Figure 2 shows the relative difference

$$R = \delta^2 N_e/\delta^2 N_e^{(0)} - 1, \tag{36}$$

of $\delta^2 N_e(P)$ found with the help of Eq. (35) and the population fluctuation dispersion $\delta^2 N_e^{(0)}(P) = 2D_{N_eN_e}/2\gamma_P$ found by integrating Eq. (35) without $\delta^2\Sigma(\omega)$. We see from Fig. 2 that $R < 1$, which means that the contribution from $\delta\hat{\Sigma}$ to the population is relatively small for $P < 2$. So, for the sake of simplicity, we drop the first term in Eq. (12c) at the low excitation and approximate

$$\delta\hat{N}_e(\omega) \approx \hat{F}_{N_e}(\omega)/(i\omega - \gamma_P). \tag{37}$$

Calculations based on the approximation (37) demonstrate our method in a simplified setting, however, approximation (37) is not a necessary part of the method. Approximation (37) considerably simplifies the calculation of the convolutions in Eqs. (31) and (34) and, in the meanwhile, shows, as we will see, the nonnegligible influence of population fluctuations on the lasing at the low excitation. Straightforward but cumbersome calculations of convolutions beyond the approximation (37) can be done with $\delta\hat{N}_e(\omega)$ satisfying Eq. (12c) and found from Eqs. (D5) of Appendix D. We leave such calculations for the future.

With the approximation (37) the spectrum of population fluctuations is

$$\delta^2 N_e(\omega) = 2D_{N_eN_e}/\big(\omega^2 + \gamma_P^2\big). \tag{38}$$

The mean photon number $n_1 = (2\pi)^{-1}\int_{-\infty}^{\infty} n_1(\omega)d\omega$ depends on the mean population N_e of the upper lasing states. N_e can be found from the energy conservation law (9) with $n = n_1(N_e)$.

V. RESULTS AND DISCUSSION

In these examples we present the results of calculations with parameters: the wavelength of the lasing transition

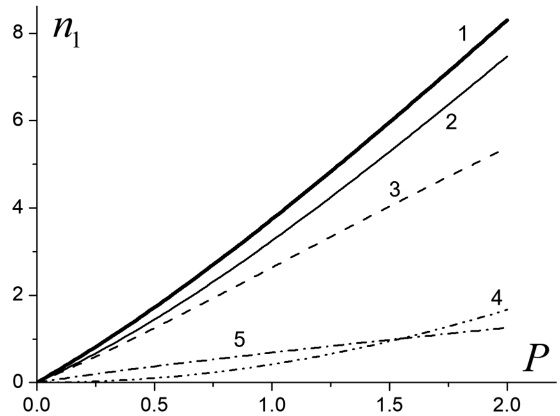

FIG. 3. The mean photon number n_1 versus the normalized pump rate P for threshold-less superradiant laser with $2\kappa/\gamma_\perp = 2$, $N_0 = 100$ resonant emitters, and nonnormalized beta factor [16] $\tilde{\beta} = 15.4 \gg 1$. Curves 1 and 2 are found with and without population fluctuations, respectively. n_1 in the curve 1 is the sum of values in curves 3, 4, and 5 taken with the same P and population inversion N. Curve 3 is due to vacuum fluctuations in the lasing mode; curves 4 and 5 are contributions of the effect of population fluctuations on stimulated and on spontaneous emission, correspondingly. The mean population inversion for curves 1, 3, 4, and 5 is smaller than for curve 2 because population fluctuations accelerate the radiation and reduce the population inversion.

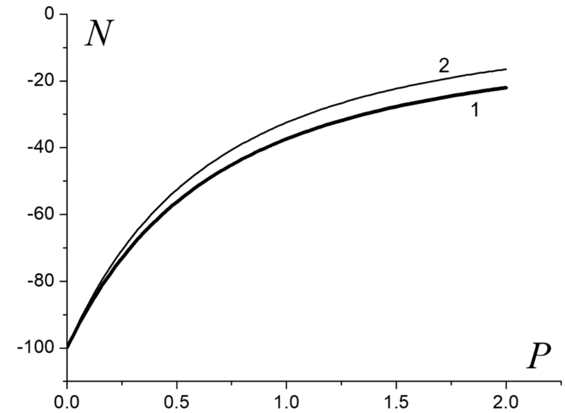

FIG. 4. The mean population inversion calculated with (curve 1) and without (curve 2) population fluctuations. Population fluctuations increase the radiation rate and deplete the population inversion. This is why curve 1 goes below curve 2.

$\lambda_0 = 1.55\ \mu$m, the background refractive index $n_r = 3.3$, the cavity mode volume $V_c = 10(\lambda_0/n_r)^3$ with $N_0 = 100$ emitters; a population relaxation rate $\gamma_\parallel = 10^9\ \mathrm{s}^{-1}$; the vacuum Rabi frequency $\Omega = (d/n_r)[\omega_0/(\varepsilon_0\hbar V_c)]^{1/2}$ with a dipole moment of the lasing transition $d = 10^{-28}$ C-m so that $\Omega = 34\gamma_\parallel$; the average atom-lasing mode-coupling factor $f = 1/2$ and the cavity quality factor $Q = 1.2 \times 10^4$ so $2\kappa = 100\gamma_\parallel$.

We vary the dephasing rate $\gamma_\perp$ and the pump P keeping all other parameters fixed. $\gamma_\perp$ is varied between 50 GHz ($2\kappa/\gamma_\perp = 2$) to 1.5 THz (with $2\kappa/\gamma_\perp = 0.07$). This is a realistic region of $\gamma_\perp$ for quantum dots [58]. We calculate the nonnormalized β-factor $\tilde{\beta} = g/\gamma_\parallel$ [16], where $g = 4\Omega^2 f/[\gamma_\perp(1 + 2\kappa/\gamma_\perp)]$ is the spontaneous emission rate into the lasing mode and the rate $\gamma_\parallel$ includes all population losses in the lasing medium. Within the chosen range for $\gamma_\perp$, $\tilde{\beta}$ varies from 15 to 1.4, so lasers with the chosen parameters have significant amounts of spontaneous emission into the lasing mode.

Similar parameters can be found in photonic crystal nanolasers with quantum-dot active media [45]; superradiant lasers with cold alkaline earth atoms [35–38], rubidium atoms [39], and quantum dots [14]. These lasers are threshold-less, with a large nonnormalized beta factor and with the significant influence of collective effects (the superradiance) [16,27,32–34]. Population fluctuations in superradiant lasers are large [16,27]. We consider the LED regime with relatively small dimensionless pump rate $P < 2$, when the mean number of the cavity photons is of the order of one or less, and when the linewidth γ_{las} of the lasing field is large $\gamma_{\mathrm{las}} > \gamma_\parallel$.

The mean photon number $n_1(P)$ for $\gamma_\perp = 50\gamma_\parallel$ is shown in Fig. 3, where we note the influence of population fluctuations on the lasing field. In Fig. 3 the bold solid curve 1 is $n_1(P)$, found in the first-order approximation with population fluctuations. The thin solid curve 2 is n_0 found without population fluctuations. The other curves are parts of n_1: curve 3 is due to fluctuations of polarization with the spectrum $n_0(\omega)$ in Eq. (29); curve 4 and curve 5 are due to the effect of population fluctuations on stimulated and on spontaneous emission, respectively; they are the integrals of spectra $n_{\mathrm{st}}(\omega)$ and $n_{sp}(\omega)$ in Eq. (29), correspondingly. Curve 1 is the sum of curves 3, 4, and 5, they depend on the same mean population inversion N found from the energy conservation law (9).

We see in Fig. 3 that population fluctuations (curves 4 and 5) give a noticeable contribution into the mean cavity photon number (curve 1). Comparing curves 1 and 2 in Fig. 3 we see that population fluctuations at the low excitation make a larger influence on the mean photon number than was predicted with the standard perturbation approach used in [16]. In Fig. 5 of [16], we see that n found with and without population fluctuations almost coincides. This is because the standard perturbation approach does not consider the influence of population fluctuations on spontaneous emission.

One can find that the population inversion N for the curve 2 is *larger* than for curves 1, 3, 4, and 5, since N is depleted, because of the population fluctuations' increase in the radiation rate; see population inversions for curves 1 (with population fluctuations) and 2 (without population fluctuations) in Fig. 4. This is why curve 3 goes below curve 2 in Fig. 3.

It is well known that the spontaneous emission is stimulated by the vacuum fluctuations of the electromagnetic field [4] and that a high density of states of the field increases the spontaneous emission rate in the cavity (Pursell effect) [59]. As an important finding, we see that the population fluctuations increase the spontaneous (and the stimulated) emission rates into the lasing mode. Such an emission rate increase may be important for highly efficient LEDs. We will estimate how large such an increase can be.

We note in Fig. 3, that the contribution of population fluctuations into spontaneous emission (curve 5) dominates the contribution into stimulated emission (curve 4) at weak pump $P < 1.5$, when the cavity photon number is small. We introduce the characteristic of the influence of the population

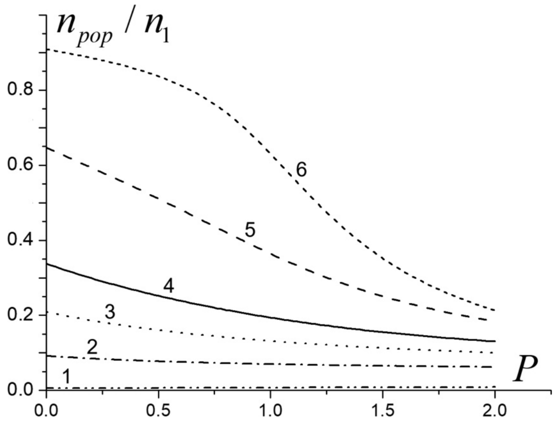

FIG. 5. The relative contribution of population fluctuations to the mean photon number for $\gamma_\perp/\gamma_\parallel = 1500$ (curve 1), 100 (2), 50 (3), 30 (4), 10 (5), and 2 (6) and other parameters the same as for Fig. 3. We see that, for the small pump, almost all photons in the lasing mode are related with population fluctuations at small $\gamma_\perp$ approaching $\gamma_\parallel$ as for curves 5 and 6.

fluctuations on the emission rate. For that we calculate the part n_{pop} of the mean number of photons

$$n_{\text{pop}} = \frac{1}{2\pi}\int_{-\infty}^{-\infty}[n_{\text{sp}}(\omega) + n_{st}(\omega)]d\omega, \tag{39}$$

caused by population fluctuations. Equation (39) is the sum of curves 4 and 5 in Fig. 3. The ratio n_{pop}/n_1 characterizes the contribution of population fluctuations into the emission rates. Smaller n_{pop}/n_1 corresponds to a smaller influence of the population fluctuations. n_{pop}/n_1 is shown in Fig. 5 as a function of the pump P for different $\gamma_\perp$. We see that n_{pop}/n_1 is reduced with P and grows for smaller $\gamma_\perp$. For curves 5 and 6 n_{pop}/n_1 is close to 1, which means that almost all photons in the lasing mode are related to population fluctuations when $P \to 0$ and for small $\gamma_\perp \to \gamma_\parallel \ll 2\kappa$. Thus we conclude that population fluctuations may considerably increase the emission rate at a weak pump in lasers with a narrow lasing transitions such that $\gamma_\perp \ll 2\kappa$. In such lasers population fluctuations are high and the collective effects are significant [16].

The limit of n_{pop}/n_1 close to 1, however, does not correspond to the perturbation approach on population fluctuations, so curves 5 and 6 in Fig. 5 must be reconsidered in higher orders of the approximation. We show curves 5 and 6 in Fig. 5 since they display a trend of the increase of the emission rate by population fluctuations, when (i) the pump P became smaller and (ii) for bad-cavity lasers, where the cavity dumping rate 2κ is relatively large $2\kappa > \gamma_\perp$. Figure 5 indicates the possibility of a high acceleration of the radiation from LEDs at a weak pump and on the corresponding increase of the LED efficiency by population fluctuations. Determining the maximum radiation rate increase at the weak pump is an interesting topic important for applications, but it is beyond the first-order perturbative scheme. We leave this topic for the future. From Fig. 5 we learn that the expected increase of the radiation rate by population fluctuations may be of the order, or even larger, than the radiation rate taken without population fluctuations.

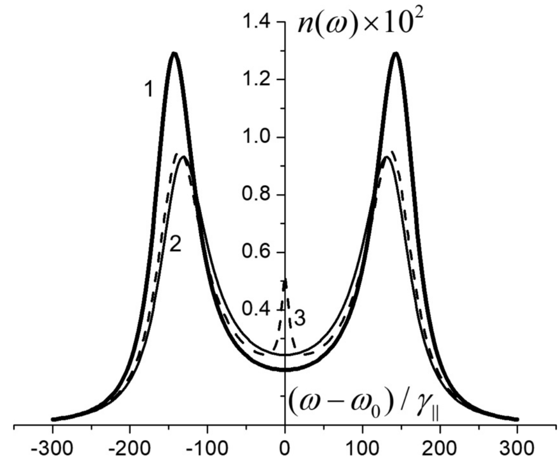

FIG. 6. Photon number spectra found with (the solid curve 1) and without (the thin curve 2) population fluctuations. $P = 1$, other parameters are the same as for Fig. 3. The dashed curve 3 is a result of [16] found with population fluctuations. The narrow peak in the center of curve 3 disappears in the present approach, while the mean photon number (the height of the spectrum) increases (compare curves 1 and 3).

Figure 6 shows spectra of the lasing field calculated with (solid curve 1) and without (thin curve 2) population fluctuations for $\gamma_\perp = 50\gamma_\parallel$ (the same as for Fig. 3) and for $P = 1$. The two peaks in spectra in Fig. 6 are because of the collective Rabi splitting [27].

According to Fig. 6, the present approach does not predict a narrow peak in the center of the spectra found in [16]. Instead we see the increase of sideband peaks due to population fluctuations. This is because the approximation $(\hat{a}\delta\hat{N}_e)_\omega \approx \sqrt{n}\delta\hat{N}_e(\omega)$ used in [16] ignores the finite width of the field spectrum and the effect of population fluctuations on the spontaneous emission into the lasing mode. It is not appropriate at the low excitation in the bad cavity lasers, where the population and the field fluctuations are large.

Thus we correct the results of [16] for the LED regime by making a more accurate description of population fluctuations. Here we use a convolution of spectra for calculating nonlinear terms in laser HLE and corrected diffusion coefficients, while in [16] the approach for a high-excitation regime was directly extended to the low-excitation LED regime.

Figure 7 shows the laser linewidth [16]

$$\gamma_{\text{las}} = \frac{2\kappa + \gamma_\perp}{\sqrt{2}}\{r - 1 + \sqrt{(r-1)^2 + r^2}\}^{1/2},$$
$$r = \frac{4\kappa\gamma_\perp}{(2\kappa + \gamma_\perp)^2}(1 - N/N_{\text{th}}), \tag{40}$$

found with (curve 1) and without (curve 2) population fluctuations. The linewidth of the laser, with population fluctuations taken into account, is larger than the linewidth of the laser where population fluctuations are neglected, so population fluctuations broaden the lasing spectrum.

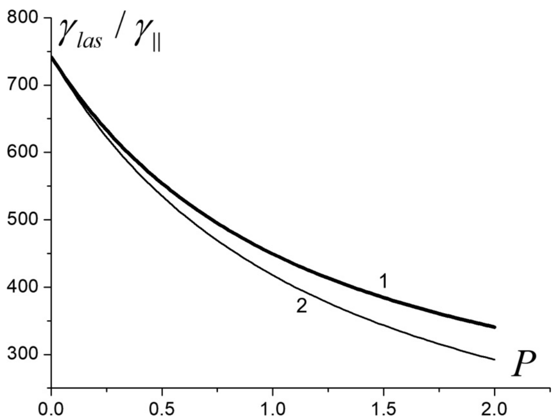

FIG. 7. Laser linewidth with (solid curve 1) and without (thin curve 2) population fluctuations for the same parameters as for Fig. 3.

VI. CONCLUSION

We consider population fluctuations as a perturbation in quantum nonlinear stochastic equations for the laser and present an approximate approach for solving such equations analytically in various orders on perturbations. As an example, we consider Maxwell-Bloch equations for the laser in the low-excitation (or LED) regime. The spectra of nonlinear terms are found as convolutions of the spectra calculated in the zero-order approximation, when population fluctuations are neglected. This approach improves the method of [16], where nonlinear terms were linearized around mean values, which is not an accurate approximation at the low excitation. Diffusion coefficients for Langevin forces are found from the requirement that Bose commutation relations for operators of the lasing field are preserved.

We find that population fluctuations accelerate spontaneous and stimulated emissions, increase the radiation rate, and, as a consequence, the mean number of lasing photons. Population fluctuations broaden the lasing spectrum. We find a larger mean photon number at the low excitation and the absence of small peaks in the center of the field spectrum shown in [16] and correct the results of [16].

Population fluctuations are high in bad cavity lasers with large gain and relatively narrow lasing transitions, such as superradiant lasers, where collective effects are significant. A large part of the radiation in the LED regime in such lasers may be related to the population fluctuations.

Lasers or LEDs with the radiation rate, increased by population fluctuations, may find applications as miniature and efficient broadband light sources.

Our approach may be applied for the theoretical analysis of various resonant systems in nonlinear and quantum optics as, for example, the optical parametric oscillator in the cavity [60].

ACKNOWLEDGMENTS

We wish to acknowledge the stimulated discussions, notes, and advice from Professor Jesper Mörk and Professor Martijn Wubs from the photonics department of the Danish Technical University.

APPENDIX A: FOURIER EXPANSION FOR OPERATORS

We consider the Fourier expansion of the Bose operator $\hat{a}(t)e^{-i\omega_0 t}$ of the lasing mode, where $\hat{a}(t)$ is changed much more slowly than $e^{-i\omega_0 t}$.

In the case of the classical field complex amplitude $a(t)$ can be represented as Fourier-integral

$$a(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} a(\omega) e^{-i\omega t} d\omega, \tag{A1}$$

where $a(\omega)$ is Fourier component of $a(t)$. Expression (A1) describes the physical fact that the electromagnetic field is a superposition of monochromatic components of different frequencies [61]. According to the Heisenberg correspondence principle [62] Fourier expansion (A1) remains true for quantum electromagnetic field, so classical variables in Eq. (A1) can be replaced by operators

$$\hat{a}(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \hat{a}(\omega) e^{-i\omega t} d\omega. \tag{A2}$$

We will come to Eq. (A2) another way, by a transition from Schrödinger to Heisenberg operators with the help of the evolution operator [63].

Suppose $|\Psi\rangle$ is a wave function of the system (of the laser in our case) and of baths interacting with the system. $|\Psi\rangle$ is, therefore, the eigenfunction of Hamiltonian H of the system and baths. In the Heisenberg representation $|\Psi\rangle$ does not depend on time. We average the operator $\hat{a}$ over $|\Psi\rangle$

$$\langle\Psi|\hat{a}(t)|\Psi\rangle = a(t). \tag{A3}$$

$a(t)$ is a random function of time because of the quantum fluctuations of the lasing mode and fluctuations due to the interaction of the mode with baths. In the stationary case $a(t)$ corresponds to the stationary random process.

Operator $\hat{a}(t)$ is related to the time-independent Schrödinger operator $\hat{a}_{\rm Sh}$ by the transformation

$$\hat{a}(t) = \exp(iHt/\hbar)\hat{a}_{\rm Sh}\exp(-iHt/\hbar), \tag{A4}$$

where $\exp(-iHt/\hbar)$ is the evolution operator [47].

Suppose, for simplicity, that $|\Psi\rangle$ can be expanded over states with discreet spectrum

$$|\Psi\rangle = \sum_{i=1}^{\infty} |\Psi_i\rangle, \tag{A5}$$

where $\{|\Psi_i\rangle\}$ is a complete set of mutually orthogonal eigenstates of Hamiltonian H.

We take a unity operator $\hat{1}$ [30,64]

$$\hat{1} = \sum_{i=1}^{\infty} |\Psi_i\rangle\langle\Psi_i|, \tag{A6}$$

and insert $\hat{1}$ into Eq. (A4) on the right and on the left sides to the operator $\hat{a}_{\rm Sh}$. After this we average Eq. (A4) over the state $|\Psi\rangle$ and come to

$$a(t) = \sum_{i,j=1}^{\infty} \langle\Psi|e^{iHt/\hbar}|\Psi_i\rangle a_{ij} \langle\Psi_j|e^{-iHt/\hbar}|\Psi\rangle, \tag{A7}$$

where $a_{ij} = \langle\Psi_i|\hat{a}_{\rm Sh}|\Psi_j\rangle$ is a matrix element of the operator $\hat{a}_{\rm Sh}$. $|\Psi_i\rangle$ are eigenfunctions of Hamiltonian H, $|\Psi\rangle$ is a superposition of states $|\Psi_i\rangle$, therefore

$$\langle\Psi|e^{iHt/\hbar}|\Psi_i\rangle = e^{iE_it/\hbar}, \quad \langle\Psi_j|e^{-iHt/\hbar}|\Psi\rangle = e^{-iE_jt/\hbar}, \quad (A8)$$

where E_i is the energy of the state $|\Psi_i\rangle$. We insert Eqs. (A8) into Eq. (A7) and come to

$$a(t) = \sum_{i,j=0}^{\infty} a_{ij} e^{-i\omega_{ij}t}, \quad (A9)$$

where $\omega_{ij} = (E_i - E_j)/\hbar$.

We consider resonant systems where the most populated states have energy close to $\hbar\omega_0$, so $\omega_{ij} \ll \omega_0$. Then we assume that matrix elements a_{ij} depend only on $E_i - E_j$, but not on E_i or E_j separately. Precisely, the dependence on $E_i \approx E_j$ is the same for relevant matrix elements taken into account. Therefore $a_{ij} = a(\omega_{ij})$. We rearrange the terms $a_{ij}e^{-i\omega_{ij}t}$ in the sum (A9) in the ascending order on ω_{ij}, use the index k instead of two indexes i and j, and rewrite Eq. (A9) as the sum over k

$$a(t) = \sum_{k=0}^{\infty} a(\omega_k) e^{-i\omega_k t}. \quad (A10)$$

Equation (A10) relates the mean $a(t)$ and matrix elements $a(\omega_k)$ of the Schrödinger operator $\hat{a}_{\rm Sh}$. Matrix elements $a(\omega_k)$ define the operator $\hat{a}(\omega_k)$, so we can rewrite the relation (A10) in terms of the operators

$$\hat{a}(t) = \sum_{k=0}^{\infty} \hat{a}(\omega_k) e^{-i\omega_k t}. \quad (A11)$$

Taking in Eq. (A11) the limit of the continuess spectrum we come to the Foruer integral (A2) for the operator $\hat{a}(t)$.

From Eq. (A4) we write

$$\hat{a}_{\rm Sh} = \exp(-iHt/\hbar)\hat{a}(t)\exp(iHt/\hbar). \quad (A12)$$

Starting with Eq. (A12) we come to the reverse Fourier transform

$$\hat{a}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \hat{a}(t) e^{i\omega t} dt, \quad (A13)$$

in a similar way as we come from Eq. (A4) to Eq. (A2).

We prefer to work with the Fourier expansions (A11) or (A2) for operators instead of the mean values as Eq. (A10). Working with these operators we can preserve the commutation relations. The expansion (A10) for means neglects the commutation relations. Obviously, $a^*(t)a(t) = a(t)a^*(t)$ while $\hat{a}^\dagger(t)\hat{a}(t) \neq \hat{a}(t)\hat{a}^\dagger(t)$. Preserving commutation relations for the field operators is important for the correct description of fluctuations at a small number of photons.

We note that there is a random function of time $a(t)$ on the left in Eq. (A10) and a random function of frequency $a(\omega_k)$ on the right in Eq. (A10). A random set of frequencies ω_k corresponds to every realization of the random process, described by $a(t)$. This way the correspondence between random processes in the time and in the frequency domains are established, for example, in numerical methods of the generation of a random signal [65]. Practically, at numerical calculations, ω_k may chose the homogeneously distributed over some interval $[-\omega_{\max}, \omega_{\max}]$, where $\omega_{\max}$ is something larger than the expected half of the maximum linewidth of the spectra of the system [65].

So each set of random frequencies corresponds to a particular realization of the random process. Such a realization may be an analog of the path integral [28,29]. Mean values of operators are the result of the averaging over many realizations.

Mean values of Fourier-component operators, for example, $\langle\hat{a}(\omega)\delta\hat{N}_e(\omega)\rangle$, are averaged over many realizations of the random processes with Fourier expansion as Eq. (A10), where a random set of frequencies is chosen for each realization.

APPENDIX B: SPECTRUM OF THE OPERATOR PRODUCT

It is sufficient to know power spectra to describe the system in the stationary state. Here we calculate the spectra of operator products approximately in the perturbation approach.

We carry out Fourier expansion of the operator $\hat{a}^\dagger$

$$\hat{a}^\dagger(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \hat{a}^\dagger(-\omega) e^{-i\omega t} d\omega, \quad (B1)$$

and take the mean $\langle\hat{a}^\dagger(t)\hat{a}(t+\tau)\rangle$. In the stationary case $\langle\hat{a}^\dagger(t)\hat{a}(t+\tau)\rangle$ does not depend on t. Therefore, if we write $\langle\hat{a}^\dagger(t)\hat{a}(t+\tau)\rangle$ with Fourier-expansions (A2) and (B1)

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \langle\hat{a}^\dagger(-\omega)\hat{a}(\omega')\rangle e^{-i(\omega+\omega')t - i\omega'\tau} d\omega d\omega', \quad (B2)$$

it must be that

$$\langle\hat{a}^\dagger(-\omega)\hat{a}(\omega')\rangle = n(\omega)\delta(\omega+\omega'). \quad (B3)$$

The physical meaning of Eq. (B3) is that there is no transition from the states of photons with different energies and $\omega \neq \omega'$ in the stationary state: the probability of such transitions, proportional to $\langle\hat{a}^+(\omega)\hat{a}(\omega')\rangle$, is zero. So the matrix of the operator $\hat{a}^\dagger(\omega)\hat{a}(\omega')$ is diagonal in the stationary state, as well as the matrices of binary products of other Fourier-component operators. This fact simplifies the calculations.

The mean number n of photons in the lasing mode is

$$n = \langle\hat{a}^\dagger(t)\hat{a}(t)\rangle = \frac{1}{2\pi} \int_{-\infty}^{\infty} n(\omega) d\omega, \quad (B4)$$

so $n(\omega)$ is a power spectrum of the lasing field.

We have seen that $n(\omega)$ is a diagonal matrix element of the operator $\hat{a}^\dagger(\omega)\hat{a}(\omega')$ in the basis $\{|\Psi_i\rangle\}$ of states of the laser and baths. Therefore,

$$dp_n(\omega) = n(\omega)d\omega/(2\pi n) \quad (B5)$$

is a probability that the lasing field is in states with energies in the interval from $\hbar(\omega_0+\omega)$ to $\hbar(\omega_0+\omega+d\omega)$. $n(\omega)/(2\pi n)$ is, therefore, a probability density.

The binary product of the Fourier-component operators $\delta\hat{N}_e(\omega)$ of the population fluctuations is

$$\langle\delta\hat{N}_e(\omega)\delta\hat{N}_e(\omega')\rangle = \delta^2 N_e(\omega)\delta(\omega+\omega'). \quad (B6)$$

Here we write $\hat{N}_e(\omega)$, not $\hat{N}_e^+(-\omega)$ [compare with Eq. (B3)] because othe population fluctuations are real quantities and $\delta\hat{N}_e^\dagger(-\omega) = \delta\hat{N}_e(\omega)$.

We consider binary products $\hat{a}(t)\delta\hat{N}_e(t)$ and $\hat{a}^\dagger(t)\delta\hat{N}_e(t)$ with zero mean $\langle\hat{a}\delta\hat{N}_e\rangle = 0$. The fact that such a mean is zero follows from Eqs. (6), when $\langle\hat{a}\rangle = 0$ and $\langle\hat{v}\rangle = 0$.

Suppose, $S_{aN_e}(\omega)$ is the spectrum of the binary products of operators $\hat{a}\delta\hat{N}_e$ We write, the same way as in Eq. (B4),

$$\langle\hat{a}^\dagger(t)\delta\hat{N}_e(t)\hat{a}(t)\delta\hat{N}_e(t)\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} S_{aN_e}(\omega)d\omega. \tag{B7}$$

We will show how $S_{aN_e}(\omega)$ is expressed through the lasing field spectrum $n(\omega)$ and the spectrum $\delta^2N_e(\omega)$ of the population fluctuations

$$\langle\delta\hat{N}_e^2(t)\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty}\delta^2N_e(\omega)d\omega. \tag{B8}$$

In follows from the analysis in Appendix A that the Fourier-component operator is expressed through the time-dependent operator by the Fourier-transform

$$(\hat{a}\delta\hat{N}_e)_\omega = \frac{1}{2\pi}\int_{-\infty}^{\infty}\hat{a}(t)\delta\hat{N}_e(t)e^{i\omega t}dt. \tag{B9}$$

Here $(\hat{a}\delta\hat{N}_e)_\omega$ is the Fourier component of $\hat{a}(t)\delta\hat{N}_e(t)$. We insert the Fourier expansions of $\hat{a}(t)$ and $\delta\hat{N}_e(t)$ into Eq. (B9) and obtain

$$(\hat{a}\delta\hat{N}_e)_\omega = \int_{-\infty}^{\infty}\hat{a}(\omega_1)\delta\hat{N}_e(\omega_2)e^{-i(\omega_1+\omega_2-\omega)t}\frac{d\omega_1 d\omega_2 dt}{(2\pi)^{3/2}}. \tag{B10}$$

We take the integral over the time in Eq. (B10) using that

$$\frac{1}{2\pi}\int_{-\infty}^{\infty}e^{-i(\omega_1+\omega_2-\omega)t}dt = \delta(\omega_1+\omega_2-\omega), \tag{B11}$$

and find

$$(\hat{a}\delta\hat{N}_e)_\omega = \int_{-\infty}^{\infty}\hat{a}(\omega_1)\delta\hat{N}_e(\omega_2)\delta(\omega_1+\omega_2-\omega)\frac{d\omega_1 d\omega_2}{(2\pi)^{1/2}}. \tag{B12}$$

Now we take the integral over $d\omega_2$ in Eq. (B12) and come to

$$(\hat{a}\delta\hat{N}_e)_\omega = \frac{1}{(2\pi)^{1/2}}\int_{-\infty}^{\infty}\hat{a}(\omega_1)\delta\hat{N}_e(\omega-\omega_1)d\omega_1. \tag{B13}$$

Therefore $(\hat{a}\delta\hat{N}_e)_\omega$ is a convolution of operators $\hat{a}(\omega)$ and $\delta\hat{N}_e(\omega)$. In a similar way we find

$$(\hat{a}^\dagger\delta\hat{N}_e)_\omega = \frac{1}{(2\pi)^{1/2}}\int_{-\infty}^{\infty}\hat{a}^\dagger(-\omega_1)\delta\hat{N}_e(\omega-\omega_1)d\omega_1. \tag{B14}$$

Now we express the mean $M = \langle\hat{a}^\dagger(t)\delta\hat{N}_e(t)\hat{a}(t)\delta\hat{N}_e(t)\rangle$ through the Fourier components of $\hat{a}^\dagger(t)$, $\hat{a}(t)$ and $\delta\hat{N}_e(t)$. First, we write

$$M = \frac{1}{2\pi}\int_{-\infty}^{\infty}\langle(\hat{a}^\dagger\delta\hat{N}_e)_{\omega_1}(\hat{a}\delta\hat{N}_e)_{\omega_2}\rangle e^{-i(\omega_1+\omega_2)t}d\omega_1 d\omega_2. \tag{B15}$$

We insert Eqs. (B13) and (B14) into Eq. (B15) and obtain

$$M = \frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\left\langle\int_{-\infty}^{\infty}\hat{a}^\dagger(-\omega_1{}')\delta\hat{N}_e(\omega_1-\omega_1{}')d\omega_1{}' \times\int_{-\infty}^{\infty}\hat{a}(\omega_1{}'')\delta\hat{N}_e(\omega_2-\omega_1{}'')d\omega_1{}''\right\rangle \times e^{-i(\omega_1+\omega_2)t}d\omega_1 d\omega_2. \tag{B16}$$

The laser at low excitation does not generate coherent radiation, $\langle\hat{a}\rangle = 0$, $\langle\hat{v}\rangle = 0$, so it follows from Eq. (6b) that $\langle\hat{a}(t)\delta\hat{N}_e(t)\rangle = 0$. Then applying the cumulant-neglected closure method [56,57] in Eq. (B16) we write

$$\langle\hat{a}^\dagger(-\omega_1{}')\delta\hat{N}_e(\omega_1-\omega_1{}')\hat{a}(\omega_1{}'')\delta\hat{N}_e(\omega_2-\omega_1{}'')\rangle \approx \langle\hat{a}^\dagger(-\omega_1{}')\hat{a}(\omega_1{}'')\rangle\langle\delta\hat{N}_e(\omega_1-\omega_1{}')\delta\hat{N}_e(\omega_2-\omega_1{}'')\rangle, \tag{B17}$$

taking into account that operators $\hat{a}$ and $\hat{a}^\dagger$ commute with $\delta\hat{N}_e$. Relation (B17) is reminiscent of the cluster expansion for correlations in the time domain [15] when

$$\langle\hat{a}^\dagger\hat{a}\delta\hat{N}_e^2\rangle \approx \langle\hat{a}^\dagger\hat{a}\rangle\langle\delta\hat{N}_e^2\rangle + 2\langle\hat{a}^\dagger\delta\hat{N}_e\rangle\langle\hat{a}\delta\hat{N}_e\rangle. \tag{B18}$$

For the laser with a low excitation the second term on the right in Eq. (B18) is zero so

$$\langle\hat{a}^\dagger\hat{a}\delta\hat{N}_e^2\rangle = \langle\hat{a}^\dagger\hat{a}\rangle\langle\delta\hat{N}_e^2\rangle. \tag{B19}$$

Equation (B17) is a "cluster expansion" for the Fourier-component operators.

According to Eqs. (B3) and (B6)

$$\begin{aligned}&\langle\hat{a}^\dagger(-\omega_1{}')\hat{a}(\omega_1{}'')\rangle\\&\quad= n(\omega_1{}')\delta(\omega_1{}'+\omega_1{}''),\\&\langle\delta\hat{N}_e(\omega_1-\omega_1{}')\delta\hat{N}_e(\omega_2-\omega_1{}'')\rangle\\&\quad=\delta^2N_e(\omega_1-\omega_1{}')\delta(\omega_1-\omega_1{}'+\omega_2-\omega_1{}'').\end{aligned} \tag{B20}$$

We insert Eq. (B20) into Eq. (B17) and Eq. (B17) into Eq. (B16) and carry out the integration in Eq. (B16) taking into account the delta functions and come to

$$\begin{aligned}M &= \frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\left(\int_{-\infty}^{\infty}n(\omega')\delta^2N_e(\omega_1-\omega_1')d\omega'\right)d\omega\\&= \frac{1}{2\pi}\int_{-\infty}^{\infty}S_{aN_e}(\omega)d\omega.\end{aligned} \tag{B21}$$

We see from Eq. (B21) that the spectrum $S_{aN_e}(\omega)$ of the operator product $\hat{a}(t)\delta\hat{N}_e(t)$

$$S_{aN_e}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty}n(\omega')\delta^2N_e(\omega_1-\omega_1')d\omega' \tag{B22}$$

is a convolution of spectra $n(\omega)$ and $\delta^2N_e(\omega)$ of operators $\hat{a}(t)$ and $\delta\hat{N}_e(t)$.

The structure of formula (B22) and the interpretation of $n(\omega)$ as a probability density [see Eq. (B5)] points out the interpretation of $S_{aN_e}(\omega)$. We calculate $\bar{S}_{aN_e} = (2\pi)^{-1}\int_{-\infty}^{\infty}S_{aN_e}(\omega)d\omega$ and, by analogy with Eq. (B5), define the probability

$$dp_{aN_e}(\omega) = S_{aN_e}(\omega)d\omega/(2\pi\bar{S}_{aN_e}). \tag{B23}$$

This is the probability of the event that an emitter and the field are in the band of states with the total energy of the emitter and the field in the interval from $\hbar(\omega_0+\omega)$ to $\hbar(\omega_0+\omega+d\omega)$, and $S_{aN_e}(\omega)/(2\pi\bar{S}_{aN_e})$ is the probability density for such an event.

Now we will comment on our perturbation approach. To find some mean value, as in the mean photon number n, we do not need to solve time-dependent equations (4) for operators. It is enough to calculate the spectrum $n(\omega)$ and use Eq. (B4). So instead of the linearization of equations of motion for operators, we approximately calculate spectra with the help of Eq. (B22). We calculate the field spectrum $n(\omega)$ neglecting by the population fluctuations, which is a zero-order approximation in the perturbation approach. The spectrum $\delta^2 N_e(\omega)$ of the population fluctuations will be found using results of the the zero-order approximation. Then, when we know $n(\omega)$ and $\delta^2 N_e(\omega)$ (though approximately), we will use Eq. (B22) for calculations of the spectrum $S_{aN_e}(\omega)$ of the operator product $\hat{a}(t)\delta\hat{N}_e(t)$. Knowing $S_{aN_e}(\omega)$ we can find from Eqs. (12a) and (12b) any spectrum and mean value in the first order on the population fluctuations and in the stationary case. The procedure may be repeated in the higher-order approximations.

To preserve the commutation relations for Bose operators of the lasing mode we calculate corrections to zero-order diffusion coefficients.

APPENDIX C: DIFFUSION COEFFICIENTS

Generalized Einstein relations [40] for the polarization of emitters lead to

$$\left\langle \frac{d}{dt}\hat{v}^\dagger\hat{v}\right\rangle = -\gamma_\perp\langle\hat{v}^\dagger\hat{v}\rangle + 2D_{v^\dagger v} = f\left\langle\frac{d}{dt}\hat{N}_e\right\rangle = f\gamma_\parallel(PN_g - N_e), \tag{C1}$$

so the diffusion coefficient

$$2D_{v^\dagger v} = f[\gamma_\perp N_e + \gamma_\parallel(PN_g - N_e)]. \tag{C2}$$

In a similar way we find

$$2D_{vv^\dagger} = f[\gamma_\perp N_g - \gamma_\parallel(PN_g - N_e)]. \tag{C3}$$

Using the energy conservation law (9) we write

$$\begin{aligned} 2D_{v^\dagger v} &= f\gamma_\perp[N_e + (2\kappa/\gamma_\perp)n],\\ 2D_{vv^\dagger} &= f\gamma_\perp[N_g - (2\kappa/\gamma_\perp)n]. \end{aligned}\tag{C4}$$

Using diffusion coefficients (C4) we calculate

$$\langle[\hat{a}_0, \hat{a}_0^\dagger]\rangle = 1 + \frac{(4\kappa/\gamma_\perp)n}{(1+2\kappa/\gamma_\perp)(N_{th}-N)}. \tag{C5}$$

So diffusion coefficients (C4) break the Bose commutation relations for $\hat{a}_0$ and they cannot be used in the zero-order approximation.

Without population fluctuations, when $\langle\frac{d}{dt}\hat{N}_e\rangle = 0$ in Eq. (C1), we have $2D^{(0)}_{v^\dagger v} = f\gamma_\perp N_e$ and $2D^{(0)}_{vv^\dagger} = f\gamma_\perp N_g$. It is shown in the main text that such zero-order diffusion coefficients preserve the commutation relations $\langle[\hat{a}_0, \hat{a}_0^\dagger]\rangle = 1$.

The sum of diffusion coefficients (C4)

$$2D_{v^\dagger v} + 2D_{vv^\dagger} = f\gamma_\perp N_0 \tag{C6}$$

does not depend on the population fluctuations. The same must be true for the sum $2D^{(1)}_{v^\dagger v} + 2D^{(1)}_{vv^\dagger}$. This is why we chose the same N_1 in diffusion coefficients (23).

APPENDIX D: EQUATIONS FOR POPULATION FLUCTUATIONS

Using Eqs. (4) and the usual rule of the differentiation of products we write equations for $\hat{\Sigma}$, given by Eq. (5), $\hat{n} = \hat{a}^\dagger\hat{a}$ and $\hat{D} = f^{-1}\sum_{i\neq j}\hat{v}_i^\dagger\hat{v}_i$. Neglecting population fluctuations we replace the population operators $\hat{N}_{e,g}$ by their means $N_{e,g}$ and obtain

$$\dot{\hat{n}} = -2\kappa\hat{n} + \Omega\hat{\Sigma} + \hat{F}_n, \tag{D1a}$$

$$\dot{\hat{\Sigma}} = -(\kappa+\gamma_\perp/2)\hat{\Sigma} + 2\Omega f(\hat{n}N + \hat{D} + N_e) + \hat{F}_\Sigma, \tag{D1b}$$

$$\dot{\hat{D}} = -\gamma_\perp\hat{D} + \Omega N\hat{\Sigma} + \hat{F}_D, \tag{D1c}$$

where $N = N_e - N_g$. Nonzero diffusion coefficients $2D_{\alpha\beta}$, $\alpha, \beta = \{n, \Sigma, D\}$ in correlations of Langevin forces $\langle\hat{F}_\alpha(t)\hat{F}_\beta(t')\rangle = 2D_{\alpha\beta}\delta(t-t')$ are

$$\begin{aligned} &2D_{nn} = 2\kappa n, \quad 2D_{\Sigma\Sigma} = f[2\kappa D + \gamma_\perp N_0 n + (2\kappa+\gamma_\perp)N_e]\\ &2D_{DD} = \gamma_\perp(N_0 D + 2N_e N_g),\\ &2D_{\Sigma n} = 2D_{n\Sigma} = \kappa\Sigma, \quad 2D_{\Sigma D} = 2D_{D\Sigma} = (\gamma_\perp/2)N_0\Sigma. \end{aligned}\tag{D2}$$

Diffusion coefficients (D2) are the same as the ones found from the generalized Einstein relations [40], apart from the term $\sim 2N_eN_g$ in $2D_{DD}$, this term must be added when we neglect population fluctuations. The derivation of diffusion coefficients (D2) will be presented in a forthcoming paper.

We separate mean values and fluctuation operators in $\hat{n}$, $\hat{\Sigma}$, and $\hat{D}$

$$\hat{n} = n + \delta\hat{n}, \qquad \hat{\Sigma} = \Sigma + \delta\hat{\Sigma}, \qquad \hat{D} = D + \delta\hat{D}, \tag{D3}$$

insert (D3) into Eqs. (D1) and obtain equations for mean values

$$0 = -2\kappa n + \Omega\Sigma, \tag{D4a}$$

$$0 = -(\kappa+\gamma_\perp/2)\Sigma + 2\Omega f(nN + D + N_e), \tag{D4b}$$

$$0 = -\gamma_\perp D + \Omega N\Sigma, \tag{D4c}$$

and for fluctuation operators $\delta\hat{n}$, $\delta\hat{\Sigma}$, and $\delta\hat{D}$

$$\delta\dot{\hat{n}} = -2\kappa\delta\hat{n} + \Omega\delta\hat{\Sigma} + \hat{F}_n, \tag{D5a}$$

$$\delta\dot{\hat{\Sigma}} = -(\kappa+\gamma_\perp/2)\delta\hat{\Sigma} + 2\Omega f(\delta\hat{n}N + \delta\hat{D}) + \hat{F}_\Sigma, \tag{D5b}$$

$$\delta\dot{\hat{D}} = -\gamma_\perp\delta\hat{D} + \Omega N\delta\hat{\Sigma} + \hat{F}_D. \tag{D5c}$$

Solving linear Eqs. (D5) by Fourier transform we obtain $\delta\hat{\Sigma}(\omega)$. With $\delta\hat{\Sigma}(\omega)$ and diffusion coefficients (D2) we find the spectrum $\delta^2\Sigma(\omega)$

$$\langle\delta\hat{\Sigma}(\omega)\delta\hat{\Sigma}(\omega')\rangle = \delta^2\Sigma(\omega)\delta(\omega+\omega'). \tag{D6}$$

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