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Spectrum of Surface Plasmons Excited by Spontaneous Quantum Dot Transitions

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Abstract—We consider quantum fluctuations of near fields of a quantum emitter (two-level system (TLS) with population inversion sustained by incoherent pumping) in the near-field zone of a plasmon (metallic) nanoparticle. The spectrum of surface plasmons excited by spontaneous transitions in the quantum emitter is obtained below the lasing threshold of such a system (spaser) in the approximation of a small number of plasmons. It is shown that the relaxation rate is the sum of the quantum emitter's rates of relaxation to its thermal reservoir and the plasmon cavity. The resulting dependence of the average number of plasmons on the pump intensity indicates the nonthreshold nature of the process.

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1. INTRODUCTION

A new branch of optics has developed in recent years, viz., quantum plasmonics [1–25], which studies the quantum regimes of the electrodynamics of plasmonic structures. The problems studied in quantum plasmonics primarily include surface-enhanced laser spectroscopy (SERS) [26, 27], near-field amplification (surface plasmon amplification by stimulated radiation emission, SPASER [28]), the development of nanosized light sources of [29–32], and the compensation of losses in plasmonic transmission lines [2, 23, 33–35] and metal materials [17, 36–41]. The latter problem is important for developing energy concentrators [42] and a superlens with a resolution exceeding the diffraction limit [37, 43, 44].

The quantum properties of plasmonic structures are manifested most clearly in a spaser [28] or in a dipole nanolaser [9], the experimental implementation of which was reported in [32, 45, 46]. A spaser can be schematically represented as a quantum-plasma system consisting of inversely excited two-level quantum dots (QDs) surrounding plasmon nanoparticles (NPs).[1] The operational principle of a spaser is analogous to that of a laser, i.e., amplification ensured by inverse population combined with the feedback produced by induced radiation of the quantum system. The condition for induced emission of radiation by an inverse quantum system in the field of the wave emitted earlier by the same system is ensured by placing the quantum system into a cavity localizing the mode being generated. The role of photons in the spaser is played by surface plasmon (SP) nanoparticles. Their localization on NPs creates conditions for the feedback. In other words, generation and amplification of near fields of NPs take place in a spaser. Amplification of SPs occurs due to nonradiative energy transfer from a QD. This process is based on the dipole–dipole [50] (or any other near-field) interaction [51] between a QD and a plasmon nanoparticle. This mechanism plays the leading role because the probability of nonradiative excitation of a plasmon is higher by a factor of $(kr_{\text{NP—TLS}})^{-3}$ than the probability of radiative de-excitation of a photon [5, 50] ($r_{\text{NP–TLS}}$ is the distance between the centers of an NP and a two-level system (QD), $k = 2\pi/\lambda$, where λ is the wavelength in vacuum). Thus, the energy transfer from the QD to the NP is effective due to the small distance between the QD and the NP.

The pumping intensity threshold value and the spaser dynamics can be described in the semiclassical approximation by the Maxwell–Bloch equations [9, 22]. In this approximation, we pass from equations for quantum operators (namely, plasmon NP annihilation operator $\hat{a}$, QD dipole transition operator $\hat{\sigma}$, and inverse population operator $\hat{D}(t) = \hat{n}_e(t) - \hat{n}_g(t)$, where $\hat{n}_e = |e\rangle\langle e|$ and $\hat{n}_g = |g\rangle\langle g|$ are the population densities of the upper and lower states of the QD) to their mean values. In this case, spontaneous radiation and quantum noise are integrated. As a consequence, the theory gives a zero-width generation line.

In [52], it was proposed that approach [53] be used for writing equations for operators $\hat{a}^{\dagger}\hat{a}$ of the number

[1] A more realistic analysis of a four-level QD does not lead to qualitatively new properties (see [47–49]).

of plasmons, upper level population density $\hat{n}_e$, and "energy exchange" operators $i(\hat{a}^\dagger\hat{\sigma} - \hat{\sigma}^\dagger\hat{a})$ instead of the Maxwell–Bloch equations and then pass to the *C*-numbers. In such an approach, additional terms appear during the derivation of the equation as a consequence of the commutation relations. To obtain a closed system of equations, correlator $\langle\hat{a}^\dagger\hat{a}\hat{n}_e\rangle = \langle\hat{a}^\dagger\hat{a}\rangle\langle n_e\rangle$ is uncoupled. The resultant equations describe the nonthreshold behavior of the spaser. In contrast to the approach based on the Maxwell–Bloch equations, the number of plasmons below the lasing threshold differs from zero. This was treated by the author of [52] as allowance for spontaneous radiation. However, this approach does not yield the spaser generation frequency or the spaser radiation linewidth.

In the recent publication [54], it was proposed that the Heisenberg–Langevin equations for plasmon NP annihilation operator $\hat{a}$, dipole transition operator $\hat{\sigma}$ in the QD, and inverse population operator $\hat{D}$ be used for describing the spaser. Then a transition was made from the operator equations to *C*-numerical equations, which were solved numerically. This approach is valid when pumping in the QD is intense and the dipole-moment amplitude for the NP is large enough to disregard quantum correlations. As should be expected, the Schawlow–Townes formula for the linewidth was obtained in a slightly modified form. However, in the vicinity of and below the spaser generation threshold, fluctuations in the quantities are on the order of magnitude of these quantities, and the approximation used in [54] is inapplicable.

Note that the problem of subthreshold dynamics in the Jaynes–Cummings model is of considerable interest in describing quantum calculations [55, 56].

In this study, we will use the approximation of a small number of plasmons to obtain the spectrum of surface plasmons excited by spontaneous transitions in a quantum emitter. It will be shown that the relaxation rate is the sum of the relaxation rates of the quantum emitter to its thermal reservoir and to the plasmon cavity. The dependence derived for the average number of plasmons on the pump intensity indicates the nonthreshold nature of the process.

2. HEISENBERG–LANGEVIN EQUATIONS OF SPASER DYNAMICS

To describe the quantum dynamics of a spaser, we can use the model Hamiltonian of the form [9, 16, 28, 38, 39, 57]

$$\hat{H} = \hat{H}_{SP} + \hat{H}_{TLS} + \hat{V}, \tag{1a}$$

where

$$\hat{H}_{SP} = \hbar\omega_{SP}\hat{\tilde{a}}^\dagger\hat{\tilde{a}}, \quad \hat{H}_{TLS} = \hbar\omega_{TLS}\hat{\tilde{\sigma}}^\dagger\hat{\tilde{\sigma}} \tag{1b}$$

are the Hamiltonians of SPs and of the two-level QD. Operator $\hat{V} = -\hat{\mathbf{d}}_{NP}\hat{\mathbf{E}}_{TLS}$, which determines the interaction between the two-level QD and the NP, can be written in the form

$$\hat{V} = \hbar\Omega_R(\hat{\tilde{a}}^\dagger + \hat{\tilde{a}})(\hat{\tilde{\sigma}}^\dagger + \hat{\tilde{\sigma}}),$$

where

$$\Omega_R = [\boldsymbol{\mu}_{NP}\boldsymbol{\mu}_{TLS} - 3(\boldsymbol{\mu}_{TLS}\cdot\mathbf{e}_r)(\boldsymbol{\mu}_{NP}\cdot\mathbf{e}_r)]/\hbar r^3$$

is the Rabi frequency and $\mathbf{e}_r$ is the unit vector of $\mathbf{r}/r$. Vectors

$$\boldsymbol{\mu}_{NP} = \sqrt{3\hbar r_{NP}^3/(\partial \mathrm{Re}\varepsilon_{NP}/\partial\omega)}\,\mathbf{e}_{NP},$$

$$\boldsymbol{\mu}_{TLS} = \mu_{TLS}\mathbf{e}_{TLS},$$

which appear in the expression for the Rabi frequency, normalize the dipole moment of the NP,

$$\hat{\mathbf{d}}_{NP} = \boldsymbol{\mu}_{NP}(\hat{\tilde{a}} + \hat{\tilde{a}}^\dagger),$$

as well as the dipole moment of the QD,

$$\hat{\mathbf{d}}_{TLS} = \boldsymbol{\mu}_{TLS}(\hat{\tilde{\sigma}}(t) + \hat{\tilde{\sigma}}^\dagger(t)).$$

Here, $\hat{\tilde{\sigma}} = |g\rangle\langle e|$ is the operator of the transition between excited $|e\rangle$ and ground $|g\rangle$ states of the QD, $\mu_{TLS} = \mathbf{r}$ is the dipole transition in the QD, and $\mathbf{e}_{NP}$ and $\mathbf{e}_{TLS}$ are the unit vectors determining the directions of the dipole moments of the NP and QD, respectively[2] (see [39] for details concerning the quantization procedure).

Assuming that the transition frequency in the QD is close to the SP frequency ($\omega_{SP} \approx \omega_{TLS}$), we will seek the solutions in the form

$$\hat{\tilde{a}}(t) \equiv \hat{a}(t)\exp(-i\omega t), \quad \hat{\tilde{\sigma}}(t) \equiv \hat{\sigma}(t)\exp(-i\omega t),$$

where $\hat{a}(t)$ and $\hat{\sigma}(t)$ are the slowly varying amplitudes. Note that the population inversion operator $\hat{D}(t)$ is "slow" by definition. Disregarding rapidly oscillating terms on the order of $\exp(\pm 2i\omega t)$ (rotating wave approximation [58]), the interaction operator $\hat{V}$ can be written in the form of the Jaynes–Cummings Hamiltonian [59]

$$\hat{V} = \hbar\Omega_R(\hat{a}^\dagger\hat{\sigma} + \hat{\sigma}^\dagger\hat{a}). \tag{1c}$$

Using the standard commutation relations $[\hat{a}, \hat{a}^\dagger] = \hat{1}$, $[\hat{\sigma}^\dagger, \hat{\sigma}] = \hat{D}$ and proceeding from Hamiltonian (1), we obtain the following Heisenberg equations of motion [9, 60] for operators $\hat{a}(t)$ and $\hat{\sigma}(t)$ as well as for population inversion operator $\hat{D}(t)$:

$$\dot{\hat{D}} = 2i\Omega_R(\hat{a}^\dagger\hat{\sigma} - \hat{\sigma}^\dagger\hat{a}), \tag{2}$$

[2] We assume that the spatial modes of the NP and the QD which specify these directions are singled out by the geometry of the problem.

$$\dot{\hat{\sigma}} = i\delta\hat{\sigma} + i\Omega_R\hat{a}\hat{D}, \quad (3)$$

$$\dot{\hat{a}} = i\Delta\hat{a} - i\Omega_R\hat{\sigma}, \quad (4)$$

where $\delta = \omega - \omega_{\rm TLS}$ and $\Delta = \omega - \omega_{\rm SP}$ are frequency "detunings."

It should be noted that system (2)–(4) describes neither pumping nor energy dissipation. These processes were taken into account in [9, 13, 22] phenomenologically by introducing the relevant terms written in the τ-approximation. This made it possible to calculate the frequency, threshold, and amplitude of spaser radiation [9, 13, 22].

To take into account dissipation consistently, it should be borne in mind that the spaser is an open quantum system. Following [11, 59, 61], we extend our analysis to the spaser surroundings with which NP and QD interact. Without loss of generality, we can assume that these are reservoirs in the form of a continuum of boson field modes, with which NP and QD interact and relax. Depending on the predominant relaxation mechanism [62], such bosons can be phonons, polaritons, surface plasmons, etc. [50]. The Hamiltonians of the reservoirs with which NPs and QDs interact can be written in the form

$$\hat{H}_{\rm NP-R} = \hbar\sum_j \omega_j \hat{b}_j^\dagger \hat{b}_j, \quad (5)$$

$$\hat{H}_{\rm TLS-R} = \hbar\sum_j \omega_j \hat{c}_j^\dagger \hat{c}_j, \quad (6)$$

where $\hat{b}$, $\hat{b}^\dagger$, $\hat{c}$ and $\hat{c}^\dagger$, are the annihilation and creation operators for bosons from the thermal reservoir for NPs and QDs, respectively, and the Hamiltonians of the interaction of NPs and QDs with these reservoirs are given by

$$\hat{V}_R = \hbar\sum_j \gamma_{j,1}(\hat{b}_j^\dagger\hat{a} + \hat{a}^\dagger\hat{b}_j) + \hbar\sum_j \gamma_{j,2}(\hat{c}_j^\dagger\hat{\sigma} + \hat{\sigma}^\dagger\hat{c}_j). \quad (7)$$

Hamiltonian $\hat{V}_R$ is also written in the rotating wave approximation because the interaction with the reservoirs is of the resonant type [59, 63]. The Hamiltonian of the total "spaser + reservoirs" system can be written in the form

$$\hat{H} = \hat{H}_{\rm SP} + \hat{H}_{\rm TLS} + \hat{V} + \hat{H}_{\rm NP-R} + \hat{H}_{\rm TLS-R} + \hat{V}_R. \quad (8)$$

Using this Hamiltonian, we can write the Heisenberg equations for the operators of the reservoir; under the assumption that the time of correlation of reservoir variables is much shorter than the characteristic time of variation of the system (Markov approximation), we obtain the following equations for the spaser [59, 63]:

$$\dot{\hat{a}} = (i\Delta - 1/\tau_a)\hat{a} - i\Omega_R\hat{\sigma} + \hat{F}_a(t), \quad (9)$$

$$\dot{\hat{\sigma}} = (i\delta - 1/\tau_\sigma)\hat{\sigma} + i\Omega_R\hat{a}\hat{D} + \hat{F}_\sigma(t), \quad (10)$$

$$\dot{\hat{D}} = 2i\Omega_R(\hat{a}^\dagger\hat{\sigma} - \hat{\sigma}^\dagger\hat{a}) - (D_0 + 1)/2\tau_\sigma + \hat{F}_D(t), \quad (11)$$

where

$$\tau_a^{-1} = \pi g_1(\omega_{\rm SP})[\gamma_{j,1}(\omega_{\rm SP})]^2, \quad (12)$$

$$\hat{F}_a(t) = -i\sum_j \gamma_{j,1}\hat{b}_j(0)e^{-i(\omega_j - \omega)t}, \quad (13)$$

$$\tau_\sigma^{-1} = \pi g_2(\omega_{\rm TLS})[\gamma_{j,2}(\omega_a)]^2, \quad (14)$$

$$\hat{F}_\sigma(t) = i\hat{D}(t)\sum_j \gamma_{j,2}\hat{c}_j(0)e^{-i(\omega_j - \omega)t}, \quad (15)$$

$$\hat{F}_D(t) = 2i\sum_j \gamma_{j,2}(\hat{c}_j^\dagger(0)e^{i(\omega_j - \omega)t}\sigma(t) - \sigma^\dagger(t)\hat{c}_j(0)e^{-i(\omega_j - \omega)t}), \quad (16)$$

and $g_{1,2}(\omega)$ is the density of states of the bosonic modes of the reservoirs with which NPs and QDs interact. Two-time correlators of operators $\hat{F}_a$, $\hat{F}_\sigma(t)$, and $\hat{F}_D(t)$ in the Markov approximation are delta functions such as [59, 63]

$$\langle \hat{F}_a^\dagger(t)\hat{F}_a(t')\rangle = \frac{2}{\tau_a}\bar{n}\delta(t - t'),$$
$$\langle \hat{F}_a(t)\hat{F}_a^\dagger(t')\rangle = \frac{2}{\tau_a}(\bar{n} + 1)\delta(t - t'), \quad (17)$$
$$\langle \hat{F}_a(t)\hat{F}_a(t')\rangle = \langle \hat{F}_a^\dagger(t)\hat{F}_a^\dagger(t')\rangle = 0,$$

and the occupation numbers $\bar{n} = (\exp(\hbar\omega/k_{\rm B}T) - 1)^{-1}$ of bosonic modes of reservoirs at optical frequencies can be disregarded because $\bar{n}_t$ is much smaller than unity even at room temperature. The pumping of QDs by external sources can be taken into account phenomenologically by addition of relaxation term $(\hat{1} - \hat{D})/\tau_p$ to formula (11), where τ_p is the pumping rate [9]. Denoting $\hat{D}_0 = (\tau_p - \tau_\sigma/2)/(\tau_p + \tau_\sigma/2)\hat{1}$ and $\tau_D^{-1} = 2\tau_\sigma^{-1} + \tau_p^{-1}$, we finally obtain

$$\dot{\hat{a}} = \left(i\Delta - \frac{1}{\tau_a}\right)\hat{a} - i\omega_R\hat{\sigma} + \hat{F}_a(t), \quad (18)$$

$$\dot{\hat{\sigma}} = \left(i\delta - \frac{1}{\tau_\sigma}\right)\hat{\sigma} + i\omega_R\hat{a}\hat{D} + \hat{F}_\sigma(t), \quad (19)$$

$$\dot{\hat{D}} = 2i\omega_R(\hat{a}^\dagger\hat{\sigma} - \hat{\sigma}^\dagger\hat{a}) - \frac{\hat{D} - \hat{D}_0}{\tau_D} + \hat{F}_D(t). \quad (20)$$

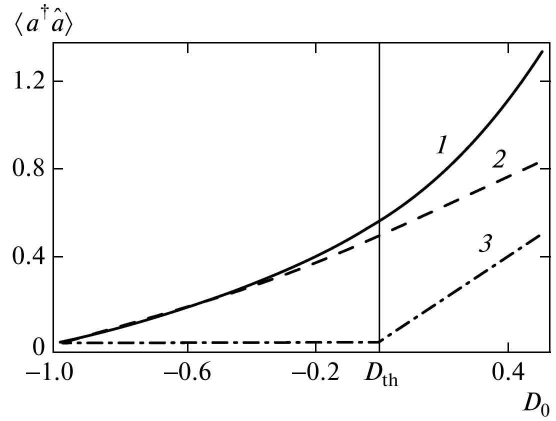

Fig. 1. Dependence of average number $\langle \hat{a}^\dagger \hat{a} \rangle$ of plasmons on pumping D_0; curve *1* corresponds to the case when quantum fluctuations are taken into account, curve *2* describes the result obtained in [52] based on uncoupling of correlator $\langle \hat{a}^\dagger \hat{a} \hat{n}_e \rangle = \langle \hat{a}^\dagger \hat{a} \rangle \langle n_e \rangle$; and curve *3* is calculated using the Maxwell–Bloch equations. The vertical line corresponds to the threshold value of pumping, obtained from the system of Maxwell–Bloch equations, $D_{\text{th}} = (\tau_a \tau_\sigma \Omega_R^2)^{-1}$. These dependences were calculated for the following values of constants: $\tau_a = 4 \times 10^{-14}$ s, $\tau_\sigma = 10^{-11}$ s, $\tau_D = 10^{-13}$ s, $\Omega_R^{-1} = 10^{-13}$ s, and $D_{\text{th}} = 0.0025$.

Equations (18)–(20) are the Heisenberg–Langevin equations for the spaser [11, 54].

3. CHAIN OF EQUATIONS FOR MEAN VALUES OF OPERATORS

System of equations (18)–(20) is of the operator type. To get an idea about the dynamics of the process, we must pass to observable quantities (mean values of these operators). However, this system is nonlinear, and Eq. (19) contains the product of operators $\hat{a}\hat{D}$, which is also an operator; to close the system, we must write the Heisenberg equation for this operator, which will contain the product of an even larger number of operators. Therefore, we obtain an infinite chain of equations describing the dynamics of the mean values of operators of physical quantities [59].

We are interested in the spaser operation near and below of the generation threshold. This means that the average number of plasmons must be small; for this reason, we will consider only two states ($|0\rangle$ and $|1\rangle$) in the subspace of states, which correspond to the absence of excited plasmons and to the state with a single excited plasmon; in other words, we assume that $\hat{a}^2|0\rangle = 0$ and $\hat{a}^{\dagger 2}|1\rangle = 0$. In this approximation, only operators $\hat{a}$, $\hat{a}^\dagger$, and $\hat{a}^\dagger\hat{a}$ (multiplied by any combinations of atomic operators) have nonzero mean values. Therefore, we will henceforth disregard the terms containing the second and higher powers of the creation and annihilation operators. For example, this concerns operators $\langle \hat{\sigma}^\dagger \hat{D} \hat{a}^2 \rangle$ and $\langle \hat{a}^\dagger (1 + \hat{D}) \hat{a}^2 \rangle$. Indeed, in this approximation, we have

$$\langle \hat{\sigma}^\dagger \hat{D} \hat{a}^2 \rangle = \text{Tr}(\hat{\sigma}^\dagger \hat{D} \hat{a}^2 \rho)$$

$$= \text{Tr}\left(\hat{\sigma}^\dagger \hat{D} \hat{a}^2 \sum_{\substack{i, i' = 0, 1 \\ j, j' = e, g}} \rho_{i, i'} |i, j\rangle\langle i', j'| \right) = 0.$$

With such an approach, two closed subsystems can be singled out from the infinite chain of equations. In our further analysis, we will also take into account the fact that the mean values of noise operators are zero:

$$\langle \hat{F}_a \rangle = \langle \hat{F}_\sigma \rangle = \langle \hat{F}_D \rangle = 0.$$

The first group of equations describes the dynamics of the mean values of Hermite operators of the number of plasmons, $\hat{a}^\dagger \hat{a}$, QD population inversion, $\hat{D}$, and energy exchange, $\hat{\sigma}^\dagger \hat{a} - \hat{a}^\dagger \hat{\sigma}$ and $\hat{a}^\dagger \hat{D} \hat{a}$:

$$\langle \dot{\hat{a}^\dagger \hat{a}} \rangle = -2\langle \hat{a}^\dagger \hat{a} \rangle / \tau_a + i\Omega_R \langle \hat{\sigma}^\dagger a - a^\dagger \sigma \rangle, \quad (21)$$

$$\langle \dot{\hat{D}} \rangle = -1/\tau_D(\langle \hat{D} \rangle - D_0) - 2i\Omega_R \langle \hat{\sigma}^\dagger a - a^\dagger \sigma \rangle, \quad (22)$$

$$\begin{aligned} i\langle \dot{\hat{\sigma}^\dagger a - a^\dagger \sigma} \rangle &= -(1/\tau_a + 1/\tau_\sigma)\langle \hat{\sigma}^\dagger a - a^\dagger \sigma \rangle \\ &+ \Omega_R \langle \hat{D} \rangle + 2\Omega_R \langle \hat{a}^\dagger \hat{D} \hat{a} \rangle + \Omega_R, \end{aligned} \quad (23)$$

$$\begin{aligned} \langle \dot{\hat{a}^\dagger \hat{D} \hat{a}} \rangle &= -(2/\tau_a + 1/\tau_D)\langle \hat{a}^\dagger \hat{D} \hat{a} \rangle \\ &+ D_0 \langle \hat{a}^\dagger \hat{a} \rangle / \tau_D - i\Omega_R \langle \hat{\sigma}^\dagger \hat{a} - \hat{a}^\dagger \hat{\sigma} \rangle. \end{aligned} \quad (24)$$

The time-independent solution to this system for $\langle \hat{a}^\dagger \hat{a} \rangle$, $\langle \hat{D} \rangle$, and $\langle \hat{a}^\dagger \hat{D} \hat{a} \rangle$ in the limit $\tau_a \ll \Omega_R^{-1}$, τ_D, τ_σ has the form

$$\langle \hat{a}^\dagger \hat{a} \rangle = (\Omega_R \tau_a)^2 (1 - \Omega_R^2 \tau_a \tau_\sigma)(1 + D_0)/2, \quad (25)$$

$$\langle \hat{D} \rangle = -1 + (1 + D_0)(1 - \Omega_R^2 \tau_a \tau_\sigma),$$

$$\langle \hat{a}^\dagger \hat{D} \hat{a} \rangle = -(\Omega_R \tau_a)^2 (1 - \Omega_R^2 \tau_a \tau_\sigma)(1 + D_0)/2. \quad (26)$$

Factor $(1 + D_0)$ indicates that for any finite occupation number of the upper level ($D_0 > -1$), the number of plasmons differs from zero and the population inversion differs from the value of D_0 specified by incoherent pumping. Note that in this analysis, we take into account quantum correlations between the atom and the field (namely, we have retained the commutation relations between operators and, as a consequence, zero fluctuation energies of the dipole moments of NPs and QDs). In the semiclassical model [9, 22–25, 52], the amplitudes dipole moments of NPs and QDs, as well as of their energies, are equal to zero (Fig. 1).

It should be noted that for the characteristic decay times in a metallic NP and a semiconducting QD ($\tau_a \sim 10^{-14}$ s, $\tau_\sigma \sim 10^{-11}$ s, $\tau_D \sim 10^{-13}$ s, and $\Omega_R^{-1} \sim 10^{-13}$ s [64–67]), the number $\langle \hat{a}^\dagger \hat{a} \rangle = (D_0 - D_{\rm th})\tau_a/4\tau_D$ of inducibly excited plasmons in the semiclassical approximation [9, 22] under experimentally attainable pump intensities [32, 45] is found to be small. However, the theory developed above cannot be used for $D_0 > D_{\rm th}$ because it exceeds the approximation used in it, predicting that $\langle \hat{a}^\dagger \hat{a} \rangle \geq 1$.

The second subsystem describes the dynamics of average non-Hermite operators $\hat{a}$ and $\hat{\sigma}$ of NP and QD dipole moments, as well as $\hat{a}\hat{D}$ and $\hat{a}\hat{\sigma}^\dagger\hat{a}$:

$$\langle \dot{\hat{a}} \rangle = \left(i\Delta - \frac{1}{\tau_a}\right)\langle \hat{a} \rangle - i\Omega_R \langle \hat{\sigma} \rangle, \tag{27}$$

$$\langle \dot{\hat{\sigma}} \rangle = \left(i\delta - \frac{1}{\tau_\sigma}\right)\langle \hat{\sigma} \rangle + i\Omega_R \langle \hat{a}\hat{D} \rangle, \tag{28}$$

$$\begin{aligned}\langle \dot{\overline{\hat{a}\hat{D}}} \rangle &= (i\Delta - (1/\tau_a + 1/\tau_D))\langle \hat{a}\hat{D} \rangle \\ &+ \langle \hat{a} \rangle D_0/\tau_D - i\Omega_R \langle \hat{\sigma} \rangle + 2i\Omega_R \langle \hat{a}^\dagger \hat{\sigma} \hat{a} \rangle,\end{aligned} \tag{29}$$

$$\begin{aligned}\langle \dot{\overline{\hat{a}^\dagger\hat{\sigma}\hat{a}}} \rangle &= (i\delta - (2/\tau_a + 1/\tau_\sigma))\langle \hat{a}^\dagger \hat{\sigma} \hat{a} \rangle \\ &+ i\Omega_R \langle \hat{a} \rangle/2 + i\Omega_R \langle \hat{a}\hat{D} \rangle/2,\end{aligned} \tag{30}$$

or, in matrix form,

$$\dot{\mathbf{x}} = M\mathbf{x}, \tag{31}$$

where $\mathbf{x} = \{\langle \hat{a} \rangle, \langle \hat{\sigma} \rangle, \langle \hat{a}\hat{D} \rangle, \langle \hat{a}^\dagger \hat{\sigma} \hat{a} \rangle\}^T$ and

$$M = \begin{pmatrix} -1/\tau_a & i\Omega_R & 0 & 0 \\ 0 & -1/\tau_\sigma & i\Omega_R & 0 \\ D_0/\tau_D & i\Omega_R & -(1/\tau_a + 1/\tau_D) & 2i\Omega_R \\ i\Omega_R/2 & 0 & i\Omega_R/2 & -(2/\tau_a + 1/\tau_a) \end{pmatrix}. \tag{32}$$

Note that the steady-state solution to this system is zero, which is a consequence of disregarding the nonlinear terms in the case of rupture of the chain of the Heisenberg equations. Therefore, this approximation fails to describe the Hopf bifurcation of the spaser and the phase transition from incoherent excitation of plasmons to the coherent excitation (i.e., lasing threshold). Nevertheless, this system carries rich information about the dynamics and the spectrum of the spaser.

Let us now analyze the solutions to system (31). We will find the eigenvalues of matrix M. These eigenvalues can be determined from the characteristic equation, which can be derived assuming that $\tau_a \ll \Omega_R^{-1}$, τ_D, τ_σ, which corresponds to the above characteristic decay times [64–67]:

$$\begin{aligned}&\lambda^4 + 4\lambda^3/\tau_a + 5\lambda^2/\tau_a^2 + 2\lambda/\tau_a^3 \\ &+ 2\Omega_R^2/\tau_a^2 + 2/(\tau_a^3\tau_\sigma) = 0.\end{aligned} \tag{33}$$

The roots of this equation under the same assumptions have the form

$$\lambda_1 \approx -2/\tau_a, \quad \lambda_{2,3} \approx -1/\tau_a \tag{34}$$

to within smaller-order terms, and

$$\lambda_4 \approx -\Omega_R^2\tau_a - \tau_\sigma^{-1} \tag{35}$$

(in fact, root $\lambda_{2,3}$ is multiple only in the given limit case). The decay rate is determined by the characteristic number with the smallest magnitude (i.e., $\Omega_R^2\tau_a + \tau_\sigma^{-1}$).

This result has a clear physical meaning. If the QD did not interact with the NP, the rate of its decay would be determined exclusively by the properties of the thermal reservoir and would be equal to τ_σ^{-1}. If it interacted only with the SP, and only the resonator mode decays, the decay rate in this case (in the limit $\tau_a \ll \Omega_R^{-1}$) would be $\Omega_R^2\tau_a$ (see [59]). In our case, the QD interacts both with the reservoir and with the NP which plays the role of the low-Q cavity in the given case, and its decay rate is determined by these two interactions. The above analysis shows that the relaxation rate in the limiting case when $\tau_a \ll \Omega_R^{-1}$ is the sum of the rates of the QD relaxation to the thermal reservoir and to the cavity with losses.

4. QUANTUM REGRESSION THEOREM. SPASER SPECTRUM

System (31) is linear, which allows us to determine the spaser spectrum using the quantum regression theorem [68, 69]. Let us formulate this theorem in the form convenient for further analysis. We consider a system described by operators

$$\hat{\mathbf{X}} = (\hat{X}_1, \hat{X}_2, \ldots, \hat{X}_n)^T$$

and experiencing the action of Markov noise ($\langle \hat{\mathbf{F}}(\tau') \rangle = 0$, $\langle \hat{\mathbf{F}}(\tau')\hat{\mathbf{F}}(\tau'')^T \rangle = 2D\delta(\tau'' - \tau')$, D is the matrix of correlators). If the operator dynamics is described by the linear equations

$$\dot{\hat{X}} = M\hat{\mathbf{X}} + \xi + \hat{\mathbf{F}}(t), \tag{36}$$

where matrix M and vector ξ, the form of which is determined by the properties of the system, are independent of time, the mean values of the two-time correlators are described by the linear system of equations

$$\begin{aligned} &\frac{\partial \langle \hat{\mathbf{X}}(t+\tau)\hat{\mathbf{X}}^T(t)\rangle}{\partial \tau} \\ &= M\langle \hat{\mathbf{X}}(t+\tau)\hat{\mathbf{X}}^T(t)\rangle + \xi\langle \hat{\mathbf{X}}^T(t)\rangle \end{aligned} \tag{37}$$

with the same matrix M and vector ξ [68, 69].

We will only consider the features important for further analysis (see [68–70] for details). We set $\xi = 0$, which can always be attained by a linear substitution of variables. Integrating system (36), we get

$$\begin{aligned} \hat{\mathbf{X}}(t+\tau) &= \exp(M\tau)\hat{\mathbf{X}}(t) + \exp(M(t+\tau)) \\ &\times \int_t^{t+\tau} d\tau' \exp(-M\tau')\hat{\mathbf{F}}(\tau'). \end{aligned} \tag{38}$$

Postmultiplying this relation by $\hat{\mathbf{X}}^T(t)$ and averaging over quantum states, we obtain

$$\begin{aligned} \langle \hat{\mathbf{X}}(t+\tau)\hat{\mathbf{X}}^T(t)\rangle &= \exp(M\tau)\langle \hat{\mathbf{X}}(t)\hat{\mathbf{X}}^T(t)\rangle \\ &+ \exp(M(t+\tau)) \int_t^{t+\tau} d\tau' \exp(-M\tau') \\ &\times \langle \hat{\mathbf{F}}(\tau')\hat{\mathbf{X}}^T(t)\rangle. \end{aligned} \tag{39}$$

The second term on the right-hand side of this equation is zero. Indeed, substituting the solution to Eq. (38) into this term, we obtain

$$\begin{aligned} &\exp(M(t+\tau)) \int_t^{t+\tau} d\tau' \exp(-M\tau')\langle \hat{\mathbf{F}}(\tau')\hat{\mathbf{X}}^T(t)\rangle \\ &= \exp(M(t+\tau)) \int_t^{t+\tau} d\tau' \exp(-M\tau')\langle \hat{\mathbf{F}}(\tau')\hat{\mathbf{X}}^T(0)\rangle \\ &+ \exp(M(t+\tau)) \int_t^{t+\tau} d\tau' \int_0^t d\tau'' \exp(-M\tau') \\ &\times \langle \hat{\mathbf{F}}(\tau')\hat{\mathbf{F}}^T(\tau'')\rangle \exp(-M^T\tau'')\exp(M^T t). \end{aligned} \tag{40}$$

Since the reservoir and the system are not correlated at the initial instant, we have

$$\langle \hat{\mathbf{F}}(\tau')\hat{\mathbf{X}}^T(0)\rangle = \langle \hat{\mathbf{F}}(\tau')\rangle\langle \hat{\mathbf{X}}^T(0)\rangle = 0,$$

and the first term on the right-hand side of Eq. (40) vanishes. Substituting the noise correlators

$$\langle \hat{\mathbf{F}}(\tau')\hat{\mathbf{F}}^T(\tau'')\rangle = 2D\delta(\tau'' - \tau')$$

into the second term of Eq. (40), we obtain

$$\begin{aligned} &\int_t^{t+\tau} d\tau' \int_0^t d\tau'' \delta(\tau'' - \tau')\exp(M(\tau - \tau')) \\ &\times 2D\exp(M^T(t - \tau'')) = 0. \end{aligned} \tag{41}$$

Indeed, the integration domain $[t, t+\tau] \otimes [0, t]$ in this formula intersects the carrier of the delta function $\delta(\tau'' - \tau')$ over a set with zero measure at point (t, t). Thus, expression (40) equals zero.

This result has a clear physical meaning. Since the noise is of the Markov type, the time of correlation between $\hat{\mathbf{X}}$ and $\hat{\mathbf{F}}$ is much shorter than all characteristic times of the problem. Consequently, correlators $\langle \hat{\mathbf{F}}(\tau')\hat{\mathbf{X}}^T(t)\rangle$ in expression (39) vanishes (see also [68–70]).

Thus, expression (40) assumes the form

$$\langle \hat{\mathbf{X}}(t+\tau)\hat{\mathbf{X}}^T(t)\rangle = \exp(M\tau)\langle \hat{\mathbf{X}}(t)\hat{\mathbf{X}}^T(t)\rangle. \tag{42}$$

Differentiating this expression with respect to τ, we obtain formula (37). It should be emphasized once again that the main conditions for fulfilling the quantum regression theorem are the linearity of initial system of operator equations (36) and the Markov nature of the noise.

At first glance, it appears that information on noise operators of the reservoir is lost upon a transition from formula (36) to (37). However, this is not true. First, according to the fluctuation–dissipation theorem [59, 61, 63, 68–70], the correlators of Markov noise determine the decay constants that appear explicitly in matrix M (as shown in Section 2; see formula (17)). Second, information on the reservoir temperature is contained in operator equations (27)–(30) in terms of occupation numbers $\bar{n}$ for reservoir modes (see [59] for details). These numbers were omitted because $\bar{n} \ll 1$ in the optical range even at room temperature.

Let us consider again a spaser operating near and below the threshold. Since its dynamics is described by linear system (31), its spectrum can be calculated using the quantum regression theorem.[3] According to the Wiener–Khinchin theorem, the spectrum $S(\omega)$ of the electric field is a Fourier transform of a 2D correlation function

$$\begin{aligned} S(\omega) &= \frac{1}{\pi}\mathrm{Re}\int_0^\infty d\tau \langle \hat{E}^{(-)}(\tau)\hat{E}^{+}(0)\rangle \\ &\times \exp(-i\omega t), \end{aligned} \tag{43}$$

[3] This approach was successfully used for calculating the resonance fluorescence spectrum [59, 68], the spectrum of a three-level system located near a metallic plane [71], etc.

where $\hat{E}^{(-)}$ and $\hat{E}^{(+)}$ are the negative- and positive-frequency branches of the electric field operator [59, 70]. The choice of such ordering of the operators corresponds to the spectrum of light emitted by the NP and QD dipoles [59, 72].

To calculate correlator $\langle \hat{E}^{(-)}(\tau)\hat{E}^{(+)}(0)\rangle$, we assume that the dipole moments of the NP and the two-level QD are parallel to the x axis and the point of observation lies on the z axis. Then the fields emitted by the NP and QD in the far-field zone are defined by the relations [59]

$$\hat{E}^{(+)}(\mathbf{r}, t) = \frac{\omega^2\langle \boldsymbol{\mu}\rangle}{c^2|\mathbf{r}|}\hat{x}\langle \hat{d}(t-|\mathbf{r}|/c)\rangle,$$
$$\hat{E}^{(-)}(\mathbf{r}, t) = \frac{\omega^2|\boldsymbol{\mu}|}{c^2|\mathbf{r}|}\hat{x}\langle \hat{d}^\dagger(t-|\mathbf{r}|/c)\rangle. \qquad (44)$$

Considering that $|\boldsymbol{\mu}|\hat{d} = |\boldsymbol{\mu}_{\rm NP}|\hat{a}$ for the NP and $|\boldsymbol{\mu}|\hat{d} = |\boldsymbol{\mu}_{\rm TLS}|\hat{\sigma}$ for the two-level QD, we obtain the radiation spectrum of the spaser in the form

$$S(\omega) \approx \mathrm{Re}\int_0^\infty d\tau \exp(-i\omega\tau)\langle(\mu_{\rm NP}\hat{a}^\dagger(\tau) + \mu_{\rm TLS}\hat{\sigma}^\dagger(\tau))(\mu_{\rm NP}\hat{a}(0) + \mu_{\rm TLS}\hat{\sigma}(0))\rangle. \qquad (45)$$

Thus, to calculate the spectrum of the system, we must know the following correlators:

$$\langle \hat{a}^\dagger(t+\tau)\hat{a}(t)\rangle, \quad \langle \hat{a}^\dagger(t+\tau)\hat{\sigma}(t)\rangle,$$
$$\langle \hat{\sigma}^\dagger(t+\tau)\hat{a}(t)\rangle, \quad \langle \hat{\sigma}^\dagger(t+\tau)\hat{\sigma}(t)\rangle.$$

Using the quantum regression theorem, we can find these correlators by solving system (37), in which

$$\hat{X}_1 = \hat{a}, \quad \hat{X}_2 = \hat{\sigma}, \quad \hat{X}_3 = \hat{a}\hat{D}, \quad \hat{X}_4 = \hat{a}^\dagger\hat{\sigma}\hat{a},$$

and M is defined in Eq. (32).

Figure 2 shows the spaser spectrum obtained by numerically solving system of equations (37). The spectrum has the form of a broad NP line with a narrow peak of much smaller width against its background.

This result has a clear physical meaning. The solution to system (37) is the sum of the exponentials of the product from multiplying eigenvalues λ of matrix M by time. The Fourier transform of such a function is the sum of the Lorentz lines, whose centers are determined by the imaginary parts $\mathrm{Im}\lambda$ of the eigenvalues, while halfwidths are determined by their real parts $\mathrm{Re}\lambda$. Eigenvalues λ are determined by expressions (34) and (35); consequently, the spectrum

$$S(\omega) \approx ((\omega-\omega_{\rm SP})^2 + 1/\tau_a^2)^{-1} + ((\omega-\omega_{\rm SP})^2 + (\Omega_R^2\tau_a^{-1} + \tau_\sigma^{-1})^2)^{-1} \qquad (46)$$

is the sum of two Lorentz lines: a broad NP line whose width is determined by the Joule loss in the metal (on the order of τ_a^{-1}) and a narrow QD line whose width is approximately equal to $\Omega_R^2\tau_a^{-1} + \tau_\sigma^{-1}$ and determined by the interaction between the NP and the QD.

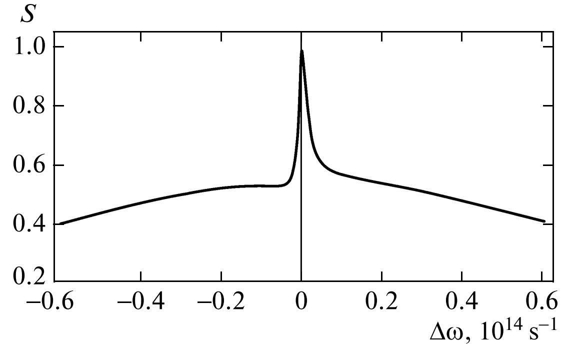

Fig. 2. Spaser spectrum below generation threshold. Parameters are the same as in Fig. 1.

Thus, the linewidth of the spaser operating near and below the threshold cannot be described by formulas of the Schawlow–Townes type, which are normally used for calculating the line width in the spectrum of quantum generators [53, 54, 58, 61] with a large number of quanta. This is due to the fact that in the regime with a small number of quanta, amplitude fluctuations of the radiation field play a significant role apart of the phase fluctuations. It was shown above that these fluctuations can be correctly taken into account using the procedure of truncation of the chain of equations (18)–(20) and the quantum regression theorem (37).

5. CONCLUSIONS

We have analyzed the operation of the spaser in the subthreshold regime. A correct calculation of the line width of the spaser operating near and below the generation threshold is based on the quantum regression theorem. This approach is applicable for $n \le 1$. It is shown that the allowance for quantum fluctuations and correlations is essential for the description of operation below the threshold, which results in a nonthreshold behavior of the spaser due to spontaneous emission of SPs by an excited QD. The equation describing the dynamics of the two-time self-correlation function $\langle(\hat{a}^\dagger(\tau) + \hat{\sigma}^\dagger(\tau))(\hat{a}(0) + \hat{\sigma}(0))\rangle$ can be obtained from system (27)–(30) for the dipole moments, which are equal to zero in the stationary state. It is important, however, that the initial condition $\langle(\hat{a}^\dagger(0) + \hat{\sigma}^\dagger(0))(\hat{a}(0) + \hat{\sigma}(0))\rangle$ for this system is determined by the set of equations (21)–(24) for the energy and dipole moments of the NP and QD, which gives a nonzero value of energies in the stationary state due to spontaneous emission.

It is shown (expression (38)) that the shape of the spaser line cannot be described by the Schawlow–Townes formula and has the form of a broad NP line serving as the background for a narrow peak associated with the QD line. The width of the peak depends on the QD linewidth (i.e., on the decay in the QD) as well as on the interaction between the NP and QD.

This analysis is extremely important in connection with heated discussions [20, 40, 47–49, 73–75] concerning the application of subthreshold spasers for compensating losses in metamaterials.

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