Intensity-dependent frequency shift in surface plasmon amplification by stimulated emission of radiation

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The operation of a surface plasmon amplification by stimulated emission of radiation- (spaser)-based nanolaser is theoretically investigated. We find that the lasing frequency undergoes a shift as the lasing intensity increases, a result that agrees with recent experiments. We show that the mechanism of the intensity-dependent shift involves a spatial deformation of the lasing mode, which is induced by the spatial hole burning in the surrounding gain media. We develop a general analytical scheme to account for the mode deformation. Our numerical calculations demonstrate good correspondence of the lasing frequency shift with the experimental data for the simplest (spherical) geometry of the spaser.

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The desire to create more compact and faster devices leads to developing new technologies and improving existing ones, such as the creation of a miniature laser. However, every laser contains a cavity whose size should not be less than half of the generated wavelength. Practically, it should be larger than this minimum even for the smallest developed lasers [1,2]. A new type of laser, proposed in Ref. [3] and experimentally demonstrated in Refs. [4–6], can overcome the aforementioned limit. The geometrical size of the resonator can be significantly reduced in comparison with the wavelength due to the usage of the surface plasmon mode instead of the resonant cavity. Thus, the metal and its surface are necessary constituents of this type of laser. Because the plasmon oscillations are coupled with gain media via the near field, the gain material should be localized in the vicinity of the metal surface to provide extra energy for the plasmon mode generation. Thus, this type of laser can be called the surface plasmon amplification by stimulated emission of radiation (spaser) [3]. This device is anticipated to become a key element in nanotechnology applications, such as (bio)-sensing [7,8], imaging [9], and information technology [10,11]. The most evident advantage of these lasers is the ability to generate coherent light with their sizes being well below the wavelength.

Different geometries were used for the experimental realization of the spasers. Spherical geometry of the metal grain was applied in Ref. [5] with the gold grain coated by the gain medium. In Ref. [4], a hybrid plasmonic waveguide consisting of a dielectric fiber made of an active medium located close to the metal surface was used. The same principles were used in the development of the spaser with metallic ring wires coated by active atoms [12].

In our paper, we theoretically investigate lasing properties of the simplest core-shell spaser's geometry in a steady-state regime [5]. The main goal of this paper is to establish the dependence of the lasing frequency on the lasing intensity, which was observed for the first time in recent experiments [4,5], although the phenomenon was not mentioned and was not discussed in the papers. Besides, there is a discrepancy between experimental and theoretical results concerning spasers. According to the theoretical paper [13], the lasing frequency should be placed between the frequency of the spontaneous emission of the active medium and the extinction maxima. However, in the experimental paper [5], the lasing frequency is redshifted relative to both of them. To reconcile this contradiction, we propose the following: The lasing frequency, indeed, lies between the extinction maxima and the spontaneous emission frequency in the threshold but it is shifted with excessing the threshold, leading to the observed phenomenon. We show the shift in the lasing frequency caused by the spatial deformation of the lasing mode due to inhomogeneous depletion of the gain medium. The quality factor of a spaser is not very large, and the depletion leads to considerable alternations in the effective dielectric permittivity of the gain medium, causing the deformation of the lasing mode.

It should be noted that the lasing frequency dependence on pumping intensity is a well-known phenomenon for the diode and some other types of lasers [14]. There are several sources of this phenomenon among which are the expansion of the cavity, variation in the refractive index, or carrier density due to the temperature increasing with the pumping. The other sources are nonlinear optical effects. The nonlinear effect considered as the source of the frequency shift in the spaser is referred to as the spatial hole burning in the gain medium [15]. It was investigated in, e.g., Ref. [16] for diode lasers.

Previous theoretical papers describing the operation of the spasers (see, e.g., Refs. [3,13,17–19]) assumed the constant spatial structure of the lasing mode. In our approach, we take into account the deformation of the lasing mode's structure with obtaining the analytical dependence of the lasing frequency on the pump intensity. Then, the results of the numerical calculations are presented. We assume the following design of the spaser: the metallic particle of radius a is coated by a shell of thickness h with embedded dye molecules, see Fig. 1. The system is illuminated by an electromagnetic wave of frequency ω_p and intensity I_p, which pumps the active media, exciting the dye molecules from ground state $|g\rangle$ to pumped state $|p\rangle$, see Fig. 1. Laser transition occurs between upper and lower laser states $|u\rangle$ and $|l\rangle$, respectively, and the frequency of the spontaneous emission is ω_{se}. We assume fast nonradiative transitions for the dye molecules from $|p\rangle$ to $|u\rangle$ and from $|l\rangle$ to $|g\rangle$ by means of phonon emission or

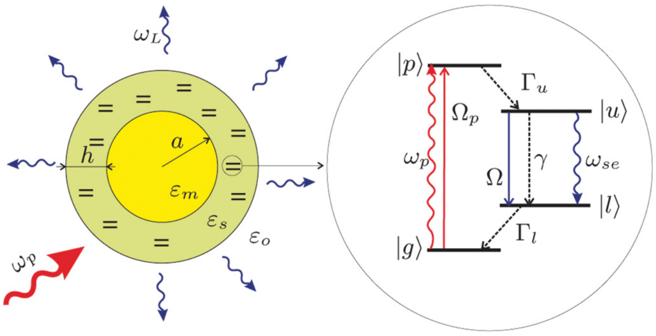

FIG. 1. (Color online) The model under study. Straight lines denote the transition rates, and wavy lines denote the frequencies of the transitions.

excitation of some other internal degrees of freedom. Note that our approach can be generalized for more complex geometries, e.g., Ref. [4]; now our aim is to uncover the main principles of the lasing frequency shift in spasers.

First, let us determine the equation describing the spatial structure of the lasing mode. The lasing frequency is ω_L, and hereinafter, we extract the fast oscillation factor from the electromagnetic field. For example, now the electric field is $\mathrm{Re}[\mathbf{E}\exp(-i\omega_L t)]$. Since the size of the system is significantly less than both the skin depth in the metal of the core and the wavelength in the material of the shell, one can neglect the curl part in the electric field $\mathbf{E}$ keeping only its potential part $\mathbf{E} = -\mathrm{grad}\,\Phi$. Divergence of Maxwell's equation $\mathrm{curl}\,\mathbf{H} = -i(\omega_L/c)\hat{\varepsilon}(\boldsymbol{r})\mathbf{E}$ is the quasistatic equation on the electric potential Φ,

$$\mathrm{div}[\hat{\varepsilon}(\boldsymbol{r})\mathrm{grad}\,\Phi] = 0, \tag{1}$$

in this approximation, where $\hat{\varepsilon}(\boldsymbol{r})$ is the local value of the dielectric permittivity of the media. Equation (1) and the condition $\Phi \to 0$ far from the spaser determine the structure of the lasing mode. The permittivity is ε_m inside the metal core and ε_o in the outer space. Inside the shell, it is $\hat{\varepsilon}_s = \varepsilon_s^{(0)} + \hat{\varepsilon}_s^a$, where $\varepsilon_s^{(0)}$ is the constant for the shell material without dye molecules and $\hat{\varepsilon}_s^a$ stems from the contribution produced by the molecules. Polarization $\mathbf{P}_a = \hat{\varepsilon}_s^a\mathbf{E}/(4\pi)$ associated with the dye molecules generally is a nonlinear function of the electric field. We describe the state of the dye molecules in terms of density matrix $\hat{\rho}$, which depends on position $\boldsymbol{r}$ and the direction of the dipole moment $\mathbf{d} = \langle u|\hat{\mathbf{d}}|l\rangle$, where $\hat{\mathbf{d}}$ is the dipole moment quantum operator. The magnitude of the dipole moment $\mathbf{d}$ is the same for all dye molecules, but its direction is random and is frozen for each molecule. The polarization is determined by the nondiagonal element $\rho_{ul} = \exp(-i\omega_L t)\rho$ of the matrix $\mathbf{P}_a(\boldsymbol{r}) = n\langle\mathbf{d}^*\rho\rangle_d$, where n is the dye molecules' concentration, the angle brackets with the low index "d" mean averaging over the random direction of the dipole moment $\mathbf{d}$, and the asterisk stands for complex conjugation. Hereinafter, the nonlinear operator $\mathrm{div}\,\hat{\varepsilon}(\boldsymbol{r})\mathrm{grad}$ is referred to as $\hat{\mathcal{H}}$ for brevity.

Second, for the emission produced by the dye molecules to be described, let us restrict ourselves to the two-level model of the system by taking only lasing states $|u\rangle$ and $|l\rangle$ into consideration. In this case, there are only three independent parameters in the truncated density matrix: the inverse population $N = \rho_{uu} - \rho_{ll}$ and the complex value of nondiagonal element ρ, whose evolution is governed by the system of equations,

$$\partial_t N = -2\,\mathrm{Im}[\Omega\rho^*] - (N - N_s)/\tau, \tag{2}$$

$$\partial_t \rho = -\Gamma_\Delta \rho - iN\Omega/2, \tag{3}$$

where $\Gamma_\Delta = \Gamma - i\Delta$ and $\Omega = (\mathbf{d}\cdot\mathbf{E})/\hbar$, thus, $|\Omega|$ is the Rabi transition rate (frequency). Equations (2) and (3) are written in rotating-wave approximation (see, e.g., Refs. [20,21]), and detuning of the light field $\Delta = \omega_L - \omega_{se}$ is assumed to be small, $\Delta \ll \omega_{se}$. The coherence relaxation rate Γ stems from homogeneous and inhomogeneous broadenings of the transition between the laser states. Population relaxation time τ, equilibrium (i.e., when the lasing mode is not excited), and inverse population N_s are determined by the pumping-wave intensity I_p. When the generation is established, one can drop temporal dependencies of N and ρ in Eqs. (2) and (3) and can find stationary values of the variables $N = N_s/(1 + \tau\Gamma|\Omega/\Gamma_\Delta|^2)$ and $\rho = -iN\Omega/(2\Gamma_\Delta)$.

Now, we take into account the pumping process and establish the dependence of the parameters τ and N_s, involved in the two-level system model (2), on intensity I_p of the pumping wave. In order to achieve this goal, we consider the more general case of the four-level system instead of the two-level system, see Fig. 1. This system can be interpreted as a two two-level subsystems; the first is the lasing subsystem with quantum states $|l\rangle$ and $|u\rangle$ (the right part in Fig. 1), and the second is the pumping subsystem with quantum states $|g\rangle$ and $|p\rangle$ (the left part in Fig. 1). These subsystems are connected to each other by fast nonradiative transitions $|p\rangle \to |u\rangle$ and $|l\rangle \to |g\rangle$ with rates Γ_u and Γ_l, respectively. Temporal equations on two nondiagonal elements ρ_{pg} and ρ_{ul} of the density matrix $\hat{\rho}$ are written in the same form (3). We assume, for the sake of simplicity, that there is no detuning in the pumping subsystem. Equations on population numbers (diagonal elements of the density matrix $\hat{\rho}$) for the lasing subsystem are now

$$\partial_t \rho_{uu} = \Gamma_u \rho_{pp} - \gamma\rho_{uu} - \mathrm{Im}[\Omega\rho^*], \tag{4}$$

$$\partial_t \rho_{ll} = -\Gamma_l \rho_{ll} + \gamma\rho_{uu} + \mathrm{Im}[\Omega\rho^*], \tag{5}$$

where $\gamma \sim \omega_{se}^3 d^2/\hbar c^3 \ll \Gamma$ is the spontaneous emission rate. We do not take into account backward transitions $|u\rangle \to |p\rangle$ and $|g\rangle \to |l\rangle$ assuming the energy difference between the absorbed and the emitted photons $\hbar(\omega_p - \omega_L)$ is much larger than the temperature of the system. Temporal equations on ρ_{pp} and ρ_{gg} can be straightforwardly restored from Eq. (4) by analogy, and the requirement for the total probability, i.e., the trace of the density matrix is unity, $\mathrm{tr}\,\hat{\rho} = 1$. The stationary solution for the four-level system leads us to the conclusion that the parameters of the two-level system considered in Eq. (2) are $N_s = 1/(1 + 2\gamma\Gamma_p/\Omega_p^2)$ and $1/\tau = 2\gamma + \Omega_p^2/\Gamma_p$, where $\Omega_p = |(\mathbf{d}_p\cdot\mathbf{E}_p)|/\hbar$ is the Rabi rate in the pumping subsystem, the corresponding dipole moment matrix element is $\mathbf{d}_p = \langle p|\hat{\mathbf{d}}|g\rangle$, $\mathbf{E}_p$ is the electric field of the pumping wave, and Γ_p is the coherence relaxation rate. Here, we assume the nonradiative rates Γ_u and Γ_l to be on the order of the coherence relaxation rates Γ and Γ_p and the moderate pumping intensities, assuming $\Omega_p \ll \Gamma$. For this case, the rates $\Gamma_{u,l}$

do not enter into the time τ. Thus, no pumping saturation is presented in our model, although the saturation can be relevant for more intense pumping. The absence of saturation in the experimental data [4,5] shows that our assumptions are reliable.

Generally speaking, N_s and τ depend on a dye molecule orientation due to the directions of the dipole moment matrix elements $\mathbf{d}_p$ and $\mathbf{d}$, which are correlated, whereas, the direction of $\mathbf{E}_p$ is fixed by the pump conditions. Thus, to calculate mean polarization $\mathbf{P}^a$ using averaging over the orientation, it is necessary to take into account this dependence. In our paper, we neglect it for simplicity in order to catch the main properties of the lasing generation in the system. Thus, we assume that the averaging over $\mathbf{d}_p$ orientation can be performed independently. For the same reason, we also neglect the spatial inhomogeneity of the gain medium inside the silica shell due to the refraction of the pumping wave near the system, assuming the pumping field amplitude E_p to be constant inside the shell. Under the made assumptions, the pumping field polarization is not relevant due to the fact that it enters in all expressions through the scalar form Ω_p.

Next, let us discuss the generation threshold for the spaser, which was found in Refs. [13,19]. The generation threshold condition can be expressed in terms of the equilibrium inverse population $N_{s,th}$ and the lasing frequency $\omega_{L,th}$ in the threshold. For this purpose, one should suppose electric field $\mathbf{E}$ of the lasing mode is small so that the inverse population N does not differ from its maximum value $N_{s,th}$. The assumption leads to the linear dependence of the polarization $\mathbf{P}_a$ on the field in Eq. (1). The result of the procedure can be represented in terms of the gain correction $\hat{\varepsilon}_s^a = -(2\pi i/3)nd^2N_s/\Gamma_\Delta$ to the dielectric permittivity of the shell. The lasing mode corresponds to the dipolelike solution of quasistatic equation (1), which exists if the relationship,

$$\frac{(\varepsilon_m + 2\varepsilon_s)(\varepsilon_s + 2\varepsilon_o)}{(\varepsilon_s - \varepsilon_m)(\varepsilon_s - \varepsilon_o)} = 2\left(1 + \frac{h}{a}\right)^{-3} \qquad (6)$$

is satisfied. Equation (6) is complex valued, and one should take into account the frequency dispersion of the core and shell permittivities ε_m and ε_s to find $N_{s,th}$ and $\omega_{L,th}$, which yields satisfaction of Eq. (6). The real part of Eq. (6) corresponds to the resonance condition, whereas, its imaginary part means balance between energy losses and pumping of the lasing mode. Using the solution, one can find the permittivity correction $\varepsilon_{s,th}^a$ and the intensity of the pumping $I_{p,th}$ in the threshold.

Equation (6) can also be used in order to find the frequency ω_{sp} of the extinction maxima for the surface plasmon when the pumping is off. The frequency ω_{sp} is the solution of Eq. (6) with the dielectric permittivity of the metal substituted by its real part. Once the frequency is found, the threshold frequency can be determined from the approximate equation [13],

$$\omega_{L,th} = \frac{\omega_{sp}/\Gamma_{sp} + \omega_{se}/\Gamma}{1/\Gamma_{sp} + 1/\Gamma}, \qquad (7)$$

where the surface plasmon resonance width is $\Gamma_{sp} = \varepsilon_m''/(\partial\varepsilon_m'/\partial\omega)$ and the value of the permittivity should be taken at surface plasmon frequency ω_{sp}. Note, the expression for Γ_{sp} accounts for only Ohmic losses. If the radiation losses are relevant, they can be considered as an additive correction to Γ_{sp}. Equation (7) implies that the lasing frequency ω_L lies between the extinction maxima ω_{sp} and the spontaneous emission frequency ω_{se}. Nevertheless, the experimental data [5] presented for the lasing frequency does not satisfy this condition. Below, we show that this disagreement can be eliminated on account of the spatial deformation of the lasing mode. The source of the lasing mode spatial deformation is the inhomogeneous depletion of the pumping (i.e., spatial hole burning, see, e.g., Ref. [15]). The effect can lead to a lasing frequency shift, for example, it was observed in Ref. [16] for diode lasers. In our paper, we demonstrate that the similar effect causes a lasing frequency shift in the spasers.

The gain correction $\hat{\varepsilon}_s^a(\boldsymbol{r})$ to the shell permittivity constant decreases with the magnitude of electric field $\mathbf{E}(\boldsymbol{r})$ and increases with pumping intensity I_p. Above the threshold, $\hat{\varepsilon}_s^a$ becomes inhomogeneous over the shell since the electric-field intensity of the lasing mode is not uniform. The stationary amplitude of the lasing mode is determined by the balance between Ohmic losses inside the metal core and pumping obtained from the gain medium. This means that the "average" over space for $\hat{\varepsilon}_s^a$ at stationary generation should be the same as at threshold point (6). First, it is convenient to establish the spatial structure of the lasing mode determining the averaging over space for $\hat{\varepsilon}_s^a$. Let us denote the electric-field potential of the mode when the pumping and the Ohmic losses inside the metal core are zero as $\Phi_{sp}(\boldsymbol{r})$. In this case, the dielectric permittivity $\varepsilon_{sp}(\boldsymbol{r})$ in the whole space is purely real, and the resonance condition (6) is satisfied. We denote the corresponding operator involved in Eq. (1) as $\hat{\mathcal{H}}_{sp}$, which is linear by definition. According to Ref. [5], we assume the quality factor Q of the spaser as a resonator to be much greater than unity. It means that both the correction $\hat{\varepsilon}_s^a$ to the shell permittivity constant and the imaginary part of the metal dielectric permittivity ε_m'' are relatively small as $1/Q$. Symmetry and self-conjugacy of the unperturbed operator $\hat{\mathcal{H}}_{sp}$ allows us to use the technique developed in quantum mechanics for perturbation theory.

The first-order correction yields to obtain the intensity of the lasing mode and the lasing frequency in the threshold. The mean-over-space value of the variance $\delta\hat{\mathcal{H}} = \hat{\mathcal{H}} - \hat{\mathcal{H}}_{sp}$ of the $\hat{\mathcal{H}}$ operator should be zero in the approximation $\int\Phi_{sp}^*\delta\hat{\mathcal{H}}\Phi_{sp}d^3r = 0$. Note that we can neglect the spatial deformation of the lasing mode structure in the first-order perturbation theory. The imaginary part of the condition provides the balance between the pumping and the dissipation in the system, whereas, the real part allows for finding the frequency of the lasing mode. The equation written particularly for the threshold gives (7). This is an expected result because the mode structure does not change being proportional to Φ_{sp}, thus, the lasing frequency should also be constant and equal to $\omega_{L,th}$. One can also treat the equation as a general condition on the amplitude of the lasing mode since $\hat{\varepsilon}_s^a$ depends on the amplitude at a given rate of pumping. Using the condition, one can evaluate both the threshold intensity of pumping $I_{p,th}$ and the dependence of the intensity of lasing mode I on the intensity of the pumping I_p (dimensionless intensities are $I_p = |\Omega_p|^2/2\gamma\Gamma_p$ and $I = |E_c|^2|\mathbf{d}|^2/2\gamma\Gamma\hbar^2$, where E_c is the electric field inside the gold core). Values I_p and I

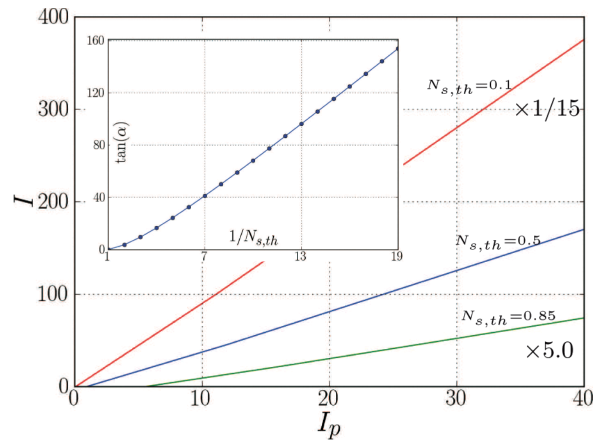

FIG. 2. (Color online) Emission intensity. Main panel: the dependence of the dimensionless stimulated emission intensity I on the dimensionless pumping intensity I_p. Inset: the tangent of the angle of the main graphic, depending on the inverse equilibrium inversion at threshold $N_{s,th}$.

are not experimentally measured intensities of the incident pumping wave and of the emitted light at the lasing frequency, respectively, but of course, they are proportional to them. In our notation, the ratio I/I_p is of the order of $(E_c/E_p)^2$. The dependence of the emission intensity on the pumping intensity was researched in detail in Ref. [19], and here, we just demonstrate the correctness of our method. We preferred to introduce these dimensionless emission and pump intensities in order to avoid using explicit values for Γ_p, $\mathbf{d}_p$, $\mathbf{d}$, and γ. We can afford to do that because the main purpose of our paper (the intensity-dependent lasing frequency shift) is separately independent of these parameters, and it only depends on such combinations as I_p and I.

The results are presented in Fig. 2. The different curves correspond to different concentrations of the dye molecules and, thus, to different threshold equilibrium inverse populations $N_{s,th}$ and threshold pumping intensities $I_{p,th}$. These two quantities are related to each other by the formula $N_{s,th} = I_{p,th}/(1 + I_{p,th})$. The dependence of the slope of the curves on threshold value $N_{s,th}$ is presented in the inset of Fig. 2, cf. with Ref. [5]. The evaluation for the zero-dimensional limit (when the spatial structure of the lasing mode has a uniform electric field over all areas containing the dye molecules, which have dipole moments directed along the field) gives $I = (\Gamma^2/|\Gamma_\Delta|^2)(1/N_{s,th} - 1)(I_p - I_{p,th})$. The numerical results were obtained for the following set of parameters: $a = 7$, $h = 15$ nm, $\varepsilon_s^0 = 2.586$, and $\varepsilon_o = 1.77$, which correspond to the experimental situation [5]. We took $\omega_{se} = 5.71 \times 10^{14}$ Hz (corresponding wavelength $\lambda_{se} = 525$ nm) and $\hbar\Gamma = 0.25$ eV ($\Gamma = 6.04 \times 10^{13}$ Hz) for Oregon Green 488, which is available on the market. The permittivity for gold is taken from Ref. [22]. In this case, we find $\omega_{sp} = 5.73 \times 10^{14}$ Hz (corresponding wavelength $\lambda_{sp} = 523$ nm) from Eq. (6) for maxima of extinction; this is in good agreement with Ref. [5]. As discussed above, our model does not account for possible pumping saturation, and it is not presented in Fig. 2 due to the fact that we consider the moderated pump intensities $E_p d_p \ll \hbar\Gamma$. Now, we can estimate the pumping field intensity corresponding to the pump saturation, evaluating $d_p \sim 10^{-17}$ esu. Thus, the saturation corresponds to the pumping field intensity of about one hundredth of the atomic field. Due to $I/I_p \sim (E_c/E_p)^2$, the ratio can be either greater or smaller than unity, see the main graph in Fig. 2. In fact, there is no contradiction with the energy conservation law in the case of $I > I_p$ since I and I_p are not experimentally measured intensities. The inset in Fig. 2 contains corresponding data.

The lasing wavelength $\lambda_{L,th} = 524$ nm found for the above-presented numerical values does not coincide with the observed lasing wavelength [5], which is redshifted and is equal to 531 nm. Now, we propose the possible source of the mismatch. As the intensity of the lasing mode grows, the pumping rate becomes nonuniform over the volume of the dielectric shell due to nonuniform depletion of the dye molecules. As a result, the spatial structure of the lasing mode undergoes a slight alternation with the intensity. The alternation results in the deviation in the lasing frequency from its threshold value $\omega_{L,th}$, now, it is $\omega_L = \omega_{L,th} + \delta_L$. To evaluate the deviation δ_L, one should develop the perturbation theory in small losses up to the second order. First, we find the correction $\delta\Phi$ for the electric-field spatial structure using the equation $\hat{\mathcal{H}}_{sp}\delta\Phi = -\delta\hat{\mathcal{H}}\Phi_{sp}$, which is valid for the first correction $\delta\Phi$ for the electric-field potential. After the correction is found, one should use the real part of the condition $\int \Phi^*\delta\hat{\mathcal{H}}\Phi\, d^3r + \int \delta\Phi^*\hat{\mathcal{H}}_{sp}\delta\Phi\, d^3r = 0$, which means that the correction up to the second order in $1/Q$ for Eq. (1) is zero. The developed method allows for determining the lasing frequency at any point of the above-threshold regime. The magnitude of the frequency shift can serve as a criterion for changing the structure of the lasing mode.

The numerical results are presented in Fig. 3. The mean-over-space depletion of the pumping is determined by the deviation in inverse population N from the undepleted level N_s. The evaluation for the zero-dimensional limit shows that the energy balance is maintained by the condition $N = N_{s,th}$. For

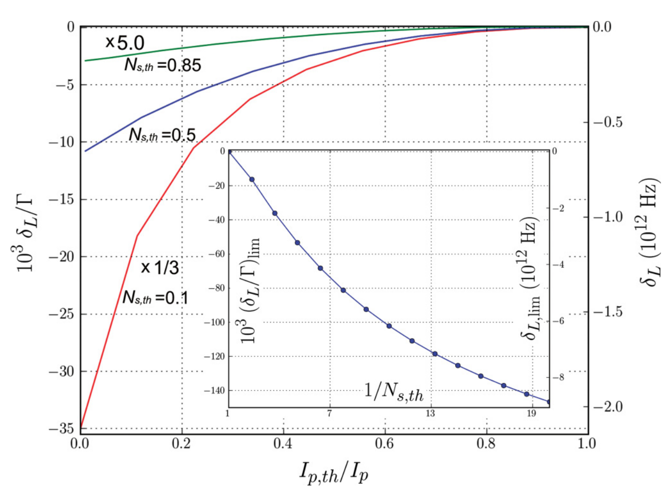

FIG. 3. (Color online) The dependence of the lasing frequency on the pump intensity. Main panel: the dependence of the lasing frequency shift δ_L from the threshold value $\omega_{L,th}$ on the inverse pump intensity normalized at the threshold. Inset: the asymptotic value of the frequency shift at the limit of strong pump intensities as a function of the inverse value of the inverse population at the threshold $1/N_{s,th}$.

this reason, we choose $1/N_{s,th}$ as the argument for the inset in Fig. 2. The experimental data of Ref. [5] suggest the existence of the frequency shift phenomenon, but this issue was not investigated in detail. For the physical parameters adopted in our calculations, the absolute value of the shift is significantly less than both the width of the spontaneous emission Γ and the width of the extinction spectra Γ_{sp}, but at the same time, it is large enough to be observed in the experiment (see the right ordinate axis in Fig. 3). The smallness is maintained by the relatively large quality factor Q of the spaser. Hence, the frequency shift produces a minor correction to the intensity of the lasing generation. One can estimate the relative correction to the lasing intensity as δ_L/Γ. This estimate validates the adopted calculation scheme in our paper, which is based on the perturbation theory in the inverse quality factor. The mode structure deformation also leads to some corrections to the lasing intensity. Since the deformation is relatively small, one can independently consider the correction to the lasing intensity and the frequency shift. This is the reason why we evaluate the magnitude of the frequency shift using the intensity of the lasing frequency obtained only on the main order.

Let us also compare the value of the intensity-dependent frequency shift with the lasing linewidth. The spectrum I_ω of the lasing mode can be found in a standard way using the model of phase diffusion [20], so the linewidth Γ_L of the spectrum can be estimated as $\Gamma_L \sim \Gamma_{sp}/n_L$ well above threshold, where n_L is the number of the surface plasmon quanta excited in the lasing mode. This means that the spectrum width becomes narrower than the secondary frequency shift for sufficiently large pumping, which was found above. So, in this case, the analyzed effect becomes significant compared with the width of the spectral line. For the chosen physical parameters, the numerical value of Γ_{sp} is close to Γ (see also the experimental results [5]). Thus, the left y axis in Fig. 3 corresponds to the ratio between the frequency shift and the linewidth within the factor, which is the number of the plasmon quanta $\delta_L/\Gamma_L \sim n_L\delta_L/\Gamma$.

In conclusion, we have developed an approach to describe the operation of spasers. By using this approach, we established the dependence of the lasing frequency on the lasing intensity. This result is interesting by itself in view of the importance of the lasing frequency and in view of its practical application. The frequency shift is associated with the change in the spatial structure of the mode. The effect arises if the Q factor of the lasing mode is not very large and the electric-field distribution of the lasing mode is nonuniform. In this case, the mode structure depends on the spatial distribution of the strength of the gain, which becomes nonuniform when the intensity of the lasing mode rises well above threshold. The approach is based on Maxwell's equations solved (in the quasistatic approximation) with the usage of perturbation theory in the inverse Q factor. All the presented results can be generalized for the arbitrary geometry of the system.

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