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Spaser in above-threshold regime: the lasing frequency shift

Parfenyev V.M., Vergeles S.S.
Moscow Institute of Physics and Technology, Moscow Region, Russia
and Landau Institute for Theoretical Physics RAS, Moscow, Russia;
e-mail: `parfenius@gmail.com, ssver@itp.ac.ru`

Operation of spaser-based nanolaser is considered theoretically. We establish that the dependence of the lasing frequency undergoes red-shift with the increase of the pumping intensity. We propose, that the mechanism leading to the red-shift is based on space deformation of the lasing mode, which is induced by inhomogeneous depletion of the gain media. We develop general analytical scheme which allows to account the mode deformation.

1 Introduction

In this work we theoretically investigate lasing properties for the simplest geometry used in [1] in a steady-state regime. The main goal of the investigation is to establish dependence of the lasing frequency on the lasing intensity. The effect can be observed for the data presented in [1, 2], although the phenomenon was not marked in the paper. Accordingly to our analysis, the shift of the lasing frequency is connected with spatial deformation of the laser working mode due to depletion of the gain media. For spaser, the quality factor is not very large, and the depletion leads to considerable alternation in effective dielectric permittivity of the gain media, that causes the deformation of the lasing mode. It should be noted, that previous theoretical works (see e.g. [3, 7, 4, 8, 9]) were built with assumption that the spatial structure of the lasing mode is invariable.

2 Investigated model

We assume the following design of the spaser: metallic particle of radius a is coated by a shell of thickness h, in which dye molecules are embedded, see Fig. 1. The system is permanently illuminated by 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, the frequency of the spontaneous emission is ω_{se}. We assume, that the dye molecules fast transit from $|p\rangle$ to $|u\rangle$ and from $|l\rangle$ to $|g\rangle$ by means of phonon emission or excitation of any other internal degrees of freedom. Note here, that our method can be generalized onto more complex geometries, e.g. [2], here our aim is to uncover the main features of the laser generation in spasers.

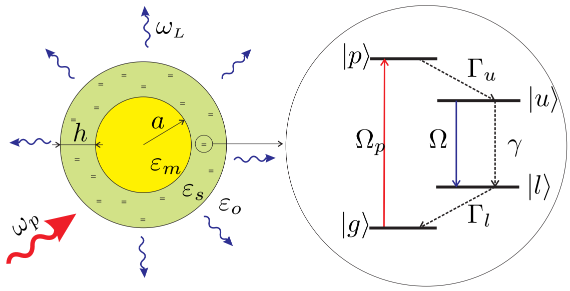

Figure 1: The model under research

Let us first determine equation, which governs spatial structure of the lasing mode. Due to the size of the system is much less than both the skin depth in the metal of the core and the wavelength in the material of the shell, one can use quasistatic equation on the electric potential Φ

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

where $\hat{\varepsilon}(\boldsymbol{r})$ is local value of dielectric permittivity of the media. Eq. (1) with the condition $\Phi \to 0$ far from the spaser determines the structure of the lasing mode. The permittivity is ε_m inside the metal core, and ε_o in 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 shell material without dye molecules and $\hat{\varepsilon}_s^a$ stems from the contribution produced by the molecules. Polarization $\mathbf{P}_a = \hat{\varepsilon}_s^a \mathbf{E}/(4\pi)$ is associated with the dye molecules, it is generally 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 matrix element $\mathbf{d} = \langle u|\hat{\mathbf{d}}|l\rangle$, 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 frozen for each molecule. The polarization is determined by non-diagonal 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 density, angle brackets with low index 'd' mean averaging over random direction of the dipole moment $\mathbf{d}$ and asterisk stands for complex conjugation. Hereinafter, nonlinear operator div$\hat{\varepsilon}(\boldsymbol{r})$grad is referred as $\hat{\mathcal{H}}$ for brevity.

To describe evolution of dye molecules, let us first restrict ourselves by two-level system model, keeping only lasing states $|u\rangle$ and $|l\rangle$ in consideration. In the case, there is only three independent parameters in the truncated density matrix, inverse population $N = \rho_{uu} - \rho_{ll}$ and complex value of non-diagonal element ρ, which evolution is governed by the system of equations

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

$$\partial_t \rho = -\Gamma_\vartriangle \rho - iN\Omega/2, \qquad (3)$$

where $\Gamma_\vartriangle = \Gamma - i\Delta$ and $\Omega = (\mathbf{dE})/\hbar$, thus $|\Omega|$ is Rabi transition rate (frequency). Equations (2, 3) are written in rotating wave approximation (see e.g. [5, 6]), detuning of the light field $\Delta = \omega_L - \omega_{se}$ is assumed to be small, $\Delta \ll \omega_{se}$. Phase decorrelation rate Γ stems from homogeneous and inhomogeneous broadening of the transition between the laser states, whereas equilibrium (i.e. at absence the field of the lasing mode) inverse population N_s and relaxation time τ are determined by the pumping wave intensity I_p. When the generation is established, one can drop temporal dependencies of N and ρ in (2), (3) and find stationary values of the variables, which are $N = N_s/(1 + \tau\Gamma|\Omega/\Gamma_\vartriangle|^2)$ and $\rho = -iN\Omega/(2\Gamma_\vartriangle)$.

Let us now establish dependence of the parameters τ and N_s involved in two-level system model (2) on intensity of pumping wave. For the purpose, we expand the two-level system up to four-level system by adding sector concerning two quantum states $|g\rangle$ and $|p\rangle$, which are brought into play in pumping process. Now we have two two-level subsystems, $|g\rangle$,$|p\rangle$ and $|l\rangle$,$|u\rangle$ which are connected with each other by fast nonradiative transitions $|p\rangle \to |u\rangle$ and $|l\rangle \to |g\rangle$ having rates Γ_u and Γ_l correspondingly. Stationary solutioin for the four-level system leads us to the conclusion that the parameters of the two-level system, considered in (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 Rabi rate in pumping sector, the corresponding dipole moment matrix element $\mathbf{d}_p = \langle p|\hat{\mathbf{d}}|g\rangle$, $\mathbf{E}_p$ is the electric field of the pumping wave and Γ_p is phase decorrelation rate. Here, we assumed the nonradiative rates Γ_u and Γ_l are order of the phase decorrelation rates Γ and Γ_p.

3 Frequency shift

The gain correction $\hat{\varepsilon}_s^a(\boldsymbol{r})$ to the shell permittivity constant decreases with magnitude of electric field $\mathbf{E}(\boldsymbol{r})$ and increases with pumping intensity I_p. Above the threshold, $\hat{\varepsilon}_s^a$ becomes nonuniform over space since the electric field intensity of the lasing mode. The stationary amplitude of lasing mode is determined by balance between Ohmic losses inside the metal core of the spaser and pumping obtained from the gain media. This means that 'average' over space value of $\hat{\varepsilon}_s^a$ at stationary generation should be the same as at threshold point. It is convenient first to establish spatial structure of the lasing mode, to determine the meaning of the 'averaging' over space for $\hat{\varepsilon}_s^a$. Let us denote $\Phi_{sp}(\boldsymbol{r})$ the electric field potential of the mode, when the pumping and the Ohmic losses inside the metal core are zero, that is when the dielectric permittivity $\varepsilon_{sp}(\boldsymbol{r})$ in whole space is pure real. We denote the corresponding operator involved in (1) as $\hat{\mathcal{H}}_{sp}$, which is liner for the case. We assume that the quality factor Q of the spaser as a resonator is much greater than unity [1]. This 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.

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 the result, the spatial structure of the lasing mode undergoes slight alternation with the intensity. The alternation results in deviation of 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 second order. First, we find the correction $\delta\Phi$ to the electric field spatial structure using the equation $\hat{\mathcal{H}}_{sp}\delta\Phi = -\delta\hat{\mathcal{H}}\Phi_{sp}$, which is valid for first correction $\delta\Phi$ for the electric field potential. After the correction is found, one should use real part of the condition $\int \Phi^* \delta\hat{\mathcal{H}}\Phi \mathrm{d}^3 r + \int \delta\Phi^* \hat{\mathcal{H}}_{sp}\delta\Phi \mathrm{d}^3 r = 0$, which means that the correction up to the second order in $1/Q$ for Eq. (1) is zero. The developed method allows to determine the lasing frequency in 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. 2. It is given for $a = 7\,\mathrm{nm}$, $h = 15\,\mathrm{nm}$, $\varepsilon_s^0 = 2.586$, $\varepsilon_o = 1.77$, the permittivity of the outer space $\varepsilon_0 = 1.77$ according to experimental work [1], $\omega_{se} = 525\,\mathrm{nm}$ and $\hbar\Gamma = 0.25\,\mathrm{eV}$. Permittivity for gold is taken from [10]. The experimental data of [1] suggests that the phenomenon of frequency shift occurs, but this issue was not investigated in details.

Let us compare the value of secondary frequency shift with the the spaser linewidth. The spectrum I_ω of the lasing mode can be found in a standard way using the model of phase diffusion [5], 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 surface plasmon quanta excited in the lasing mode and $\Gamma_{sp} = \varepsilon''_{\mathrm{m}}/(\partial\varepsilon'_{\mathrm{m}}/\partial\omega)$. This means that the spectrum width becomes less than the secondary frequency shift for sufficiently large pumping, which is found above. So, in this case the analyzed effect becomes significant compared with the width of the spectral line.

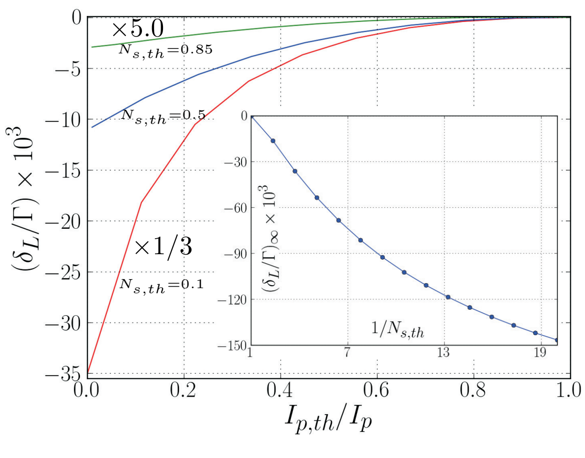

Figure 2: **The dependence of the lasing frequency on the pump intensity.** Main panel, the dependence of the lasing frequency shift δ_L from the threshold value $\omega_{L,th}$ on the inverse pump intensity normalized to the threshold. On the inset, the asymptotic value of the frequency shift at the limit of strong pump intensities as a function of inverse value of the inverse population at the threshold, $1/N_{s,th}$.

4 Conclusion

Thus, the paper presents a theoretical framework describing spaser. All results can be obtained for arbitrary geometry of the system. We have developed a new approach to the problem of spaser in above-threshold regime. The key idea is to solve Maxwell's equations in the quasistatic approximation using perturbation theory approach with the help of quantum-mechanical formalism. In particular, we found that the shift of the lasing frequency is associated with change in the structure of the mode. This result is interesting by itself, in view of the importance for the frequency of radiation, and in view of its practical application. 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 the case mode structure depends on the distribution of the pumping, which becomes nonuniform when the intensity of the lasing mode rises well above threshold.

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