NARROWING THE LINEWIDTH OF A PLASMONIC NANOLASER WITH AN INCREASE OF COUPLING BETWEEN A TWO-LEVEL SYSTEM AND A PLASMONIC NANOPARTICLE

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> Received: 25.12.2023 Accepted: 29.12.2023 Published: 18.01.2024

Abstract

The emission of a plasmonic nanolaser built from a quantized open resonator and atom is studied in the strong quantum regime when the number of excited levels in the resonator is of the order of unity. Due to the comparable size of the atom and resonator we should consider the Purcell factor for both the atom and the resonator. It is shown that the coupling of the open nanoresonator with an atom may decrease the rate of nanolaser radiation and therefore cause narrowing of the resonator linewidth.

Keywords: nanolaser, two-level system, plasmonic nanoparticle, open cavity, Purcell factor

EDN EDQPWF

doi:10.24412/2949-0553-2023-68-4-11

1. Introduction

Due to recent developments of nanoplasmonics one arises a wide range of problems associated with the new methods of spectroscopy (SERS [1,2], TERS [3,4], SNOM [5–9]), the creation of nanoscale coherent electromagnetic field sources (dipole nanolasers [10,11], nanoscale light sources [12–15], spasers [10,16–19] and the study of active metamaterials [20–24]. Underlying all these problems is the spontaneous emission of an atom in a complex environment.

The pioneering work of Purcell [25] lays the foundation of investigation of environment influence on atomic spontaneous emission (see [26] and references therein).

In contrast to Fabry-Perot resonance in usual lasers plasmonic systems operate due to plasmonic resonance. In such case the mode volume is determined by the volume of near fields modes and it may be equal several tens of nanometers. Frequently the volume of such resonator is comparable with the volume of quantum system. Thus the anti-symmetrical situation described above (big resonator - small atom) changes. One may consider inverse situation where we have small resonator and big atom. Plasmonic nanostructures may play the role of such small resonator and quantum dot may play the role of big atom. So the well-known and intuitively understandable concept of Purcell factor may be expand and be applied to resonator. Such Purcell factor describes the change of nanoresonator quality and as a consequence its linewidth. In other words there is the problem concerning the influence of intrinsic structure of resonator on its properties. Below we show that two-level quantum system (for example, quantum dot) may decrease the rate of dissipation of open nanoresonator and narrowing of nanoresonator line.

2. The classical analysis of two coupled oscillators

Firstly we consider the problem concerning the interaction of two classical oscillators and introduce classical analogue of Purcell factor.

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The dynamics of two interacting oscillators is described by the following system of equations:

$$\ddot{x}_1 + 2\gamma_1 \dot{x}_1 + \omega_1^2 x_1 = k \left(x_2 - x_1 \right), \tag{1}$$

$$\ddot{x}_2 + 2\gamma_2 \dot{x}_2 + \omega_2^2 x_2 = k \left(x_1 - x_2 \right).$$
⁽²⁾

Below we will suppose that the oscillator eigenfrequencies are much smaller than other parameters, $\omega_{1,2} \gg \gamma_{1,2}, k^{1/2}$. Introducing the new variables $\tilde{x}_1 = x_1 + i\dot{x}_1/\omega_1$, $\tilde{x}_2 = x_2 + i\dot{x}_2/\omega_2$, $\Omega = k/(\omega_1 + \omega_2)$ we obtain the following system of complex equations:

$$\frac{d}{dt}\tilde{x}_1 = \left(-i\omega_1 - \gamma_1\right)\tilde{x}_1 - i\Omega\tilde{x}_2,\tag{3}$$

$$\frac{d}{dt}\tilde{x}_2 = (-i\omega_2 - \gamma_2)\tilde{x}_2 - i\Omega\tilde{x}_1.$$
(4)

We suppose that the oscillator frequencies coincide, $\omega_1 = \omega_2$, but the first one decays much faster than the other, $\gamma_1 \gg \gamma_2, \Omega$. We consider the solution of this system with two initial conditions.

The first one corresponds to the excitation of the low-Q oscillator, $x_1(0) = 1$, $x_2(0) = 0$. In this case the dynamics of the first oscillator takes the form:

$$x_1(t) = \frac{\gamma_2 + \lambda_1}{\lambda_1 - \lambda_2} \exp\left(-i\omega + \lambda_1\right) t - \frac{\gamma_2 + \lambda_2}{\lambda_1 - \lambda_2} \exp\left(-i\omega + \lambda_2\right) t \tag{5}$$

where $\lambda_{1,2}$ are the roots of the characteristic equation

$$\lambda^2 + \lambda \left(\gamma_1 + \gamma_2\right) + \Omega^2 = 0. \tag{6}$$

Under condition $\gamma_1 \gg \gamma_2, \Omega$ we have $\lambda_1 \approx \gamma_1$, and the solution (5) takes the form:

$$x_1(t) \approx \exp\left(-i\omega - \gamma_1\right)t,\tag{7}$$

i.e. we have exponential decay with the rate γ_1 . Thus, the dumping rate does not change. This result has clear physical interpretation: high-quality resonator does not influence on the low quality one (of course, if $\gamma_1 \gg \gamma_2, \Omega$).

Now consider the opposite case, when the high quality oscillator is excited at the initial time, i.e. the initial condition $x_1(0) = 0$, $x_2(0) = 1$. In such case the dynamics of the high-quality oscillator takes the form:

$$x_2(t) \approx \exp\left(-i\omega - \Omega^2/\gamma_1\right)t,$$
(8)

e.g. we have exponential decay with the rate Ω^2/γ_1 . The ratio of this decay rate to the rate of free decay is

$$P = \Omega^2 / \gamma_1 \gamma_2, \tag{9}$$

and play the role of classical analogue of the Purcell factor.

This change in the classical case has a very clear interpretation: low-Q oscillator is connected to a high-Q oscillator and causes the change of its decay rate.

So, when two oscillators with different qualities interact with each other the decay rate of high-quality oscillator changes while the decay rate of low-quality oscillator does not change. The above case describes a classical analogue of the change of the relaxation rate of the atom during the transition from free space to the resonator (the ratio of these rates is a Purcell factor). Atom is considered to be a high-Q oscillator, moreover, its internal losses are usually completely neglected and resonator is considered as a low-Q oscillator.

This result remains valid in quantum case too, where the resonator mode plays the role of low-quality oscillator and two-level quantum system plays the role of high-quality oscillator. Here we consider the same limiting case, namely, $\gamma_a \gg \gamma_D$, Ω_R , where γ_a is the resonator relaxation rate, γ_D is the atom relaxation rate, and Rabi frequency Ω_R describes the strength of their interaction.

We consider the two-level quantum dot (QD) with ground state $|g\rangle$ and excited state $|e\rangle$ with transition being in the resonance with the dipole mode of plasmonic nanoparticle (NP). Population inversion is described by the operator $\hat{\sigma}_z = |e\rangle \langle e| - |g\rangle \langle g|$, while QD dipole moment is $\hat{\sigma}$. Plasmon electric near-field operator is proportional to plasmon annihilation operator \hat{a} while number of quanta is determined by operator $\hat{a}^+\hat{a}$. Interaction between NP and QD is dipole-dipole one, so $\hat{V} \sim \hbar \Omega_R (\hat{a}^+ \hat{\sigma} + \hat{\sigma}^+ \hat{a})$. When TLS is placed in the closed damped cavity the TLS may excite only modes of this cavity. System of equations describing dynamics of resonator mode interacting with two-level QD in the strong quantum limit when plasmon number being of the order of unity is as follows [27, 28]:

$$\frac{d}{dt}\left\langle \hat{a}^{+}\hat{a}\right\rangle = -\gamma_{a}\left\langle \hat{a}^{+}\hat{a}\right\rangle + i\Omega_{R}\left\langle \hat{\sigma}^{+}a - a^{+}\sigma\right\rangle,\tag{10}$$

$$\frac{d}{dt}\left\langle\hat{\sigma}_{z}\right\rangle = -\gamma_{D}\left(\left\langle\hat{\sigma}_{z}\right\rangle + 1\right) - 2i\Omega_{R}\left\langle\hat{\sigma}^{+}a - a^{+}\sigma\right\rangle,\tag{11}$$

$$\frac{d}{dt}i\left\langle\hat{\sigma}^{+}a-a^{+}\sigma\right\rangle = -\left(\gamma_{a}+\gamma_{D}\right)\left\langle\hat{\sigma}^{+}a-a^{+}\sigma\right\rangle/2 + \Omega_{R}\left\langle\hat{\sigma}_{z}\right\rangle + 2\Omega_{R}\left\langle\hat{a}^{+}\hat{\sigma}_{z}\hat{a}\right\rangle + \Omega_{R},\tag{12}$$

$$\frac{d}{dt}\left\langle \hat{a}^{+}\hat{\sigma}_{z}\hat{a}\right\rangle = -\left(\gamma_{a}+\gamma_{D}\right)\left\langle \hat{a}^{+}\hat{\sigma}_{z}\hat{a}\right\rangle - i\Omega_{R}\left\langle \hat{\sigma}^{+}\hat{a}-\hat{a}^{+}\hat{\sigma}\right\rangle.$$
(13)

Now, we consider the QD's behavior in empty cavity (e.g. the number of quanta is zero). For this purpose we choice the following initial conditions $\langle \hat{a}^+ \hat{a} \rangle_{t=0} = \langle \hat{a}^+ \hat{\sigma}_z \hat{a} \rangle_{t=0} = \langle \hat{\sigma}^+ a - \hat{a}^+ \hat{\sigma} \rangle_{t=0} = 0$, $\langle D \rangle_{t=0} = 1$. We are interested in spontaneous emission of QD population inversion to cavity mode. Solving equations (10) - (13) by Laplace transform we obtain:

$$\begin{aligned} \langle \hat{\sigma}_{z}(t) \rangle &= -1 + \frac{4 \exp(-(\gamma_{a} + \gamma_{D})t)}{(\gamma_{a} - \gamma_{D})^{2} - 16\Omega_{R}^{2}} \left\{ - \left(4\Omega_{R}^{2} + \gamma_{a}\gamma_{D}\right) + \right. \\ &+ \left(\left(\gamma_{a} + \gamma_{D}\right)^{2} / 4 - 2 \left(\Omega_{R}^{2} + \gamma_{a}\gamma_{D} / 4\right) + \left(\gamma_{a} + \gamma_{D}\right) \sqrt{\left(\gamma_{a} - \gamma_{D}\right)^{2} - 16\Omega_{R}^{2}} / 4 \right) \times \\ &\times \exp\left(\sqrt{\left(\gamma_{a} - \gamma_{D}\right)^{2} - 16\Omega_{R}^{2}} t / 2 \right) + \\ &+ \left(\left(\gamma_{a} + \gamma_{D}\right)^{2} / 4 - 2 \left(\Omega_{R}^{2} + \gamma_{a}\gamma_{D} / 4\right) - \left(\gamma_{a} + \gamma_{D}\right) \sqrt{\left(\gamma_{a} - \gamma_{D}\right)^{2} - 16\Omega_{R}^{2}} / 4 \right) \times \\ &\times \exp\left(- \sqrt{\left(\gamma_{a} - \gamma_{D}\right)^{2} - 16\Omega_{R}^{2}} t / 2 \right) \right\}. \end{aligned}$$
(14)

In this case under condition $\gamma_a \gg 2\Omega_R \gg \gamma_D$ decreasing of population inversion in the case of may be written as

$$\langle \hat{\sigma}_z(t) \rangle = -1 + \exp\left(-\left(4\Omega_R^2 + \gamma_a \gamma_D\right) t / (\gamma_a + \gamma_D)\right),\tag{15}$$

e.g. atom exponentially decay with the rate

$$\gamma_D^{tot} = \left(4\Omega_R^2 + \gamma_a \gamma_D\right) / \left(\gamma_a + \gamma_D\right),\tag{16}$$

(in the cases of $\gamma_a \gg \gamma_D \gg 2\Omega_R$ or $\gamma_D \gg \gamma_a$ atom exponentially decay with the rate γ_D). Wherein increasing of the atomic decay rate connects with additional joule losses in resonator due to excitation of resonator mode. Then the Purcell factor may be defined as the ration of the line width of TLS placed in resonator to the one of the TLS placed in free space

$$P_D = \frac{4\Omega_R^2 + \gamma_a \gamma_D}{(\gamma_a + \gamma_D) \gamma_D},\tag{17}$$

that in the limit $\gamma_a \gg 2\Omega_R \gg \gamma_D$ gives

$$P_D = \frac{4\Omega_R^2}{\gamma_a \gamma_D},\tag{18}$$

where factor 4 arises due to in quantum case we choose dissipation rate for energy variables.

3. Effective change of the resonator quality factor in the presence of a two-level system. Purcell factor of the resonator

As shown in the previous section, interaction of two oscillators may significantly change the relaxation rate of only high-Q one. Characteristic value of relaxation rates of open nanoresonators is the order of $\gamma_a \sim 10^{12} - 10^{14} s^{-1}$ while for quantum dots this value is the order of $\gamma_D \sim 10^9 - 10^{12} s^{-1}$. So, open nanoresonator has always smaller quality factor than quantum system and it would be seem that it is not possible to change its relaxation.

Note that in the open nanoresonator losses consist of radiative (connecting with excitation of free space modes) and non-radiative (connecting with thermal losses) ones. Excitation of near field mode (in subsequent consideration only dipole mode) is not considered as source of non-radiative losses because this

mode excitation is of interest. Thus, $\gamma_a = \gamma_a^{rad} + \gamma_a^{nonrad}$ and we can write the equation of energy relaxation in the form of

$$\frac{dW}{dt} = -\gamma_a W = -\left(\gamma_a^{rad} + \gamma_a^{nonrad}\right) W,\tag{19}$$

which yields

$$V(t) = W_0 \exp\left(-\left(\gamma_a^{nonrad} + \gamma_a^{rad}\right)t\right).$$
⁽²⁰⁾

Here W_0 is the initial energy in the resonator. Then intensity of losses is equal to

V

$$I(t) = \frac{dW}{dt} = \left(\gamma_a^{rad}W_0 + \gamma_a^{nonrad}W\right) \exp\left(-\left(\gamma_a^{nonrad} + \gamma_a^{rad}\right)t\right) = I_{rad}(t) + I_{nonrad}(t), \quad (21)$$

where $I_{rad}(t) = \gamma_a^{rad} W_0 \exp\left(-\left(\gamma_a^{rad} + \gamma_a^{nonrad}\right)t\right)$ is the intensity of irradiation into free space and $I_{nonrad}(t) = \gamma_a^{nonrad} W_0 \exp\left(-\left(\gamma_a^{rad} + \gamma_a^{nonrad}\right)t\right)$ is the intensity of non-radiative losses.

Thus the time dependence of radiation intensity is determined by the total rate of dissipation $\gamma_a^{rad} + \gamma_a^{nonrad}$, but the amplitude is determined by the radiation rate only, $I_{rad} \sim \gamma_a^{rad}$.

Now we return to the question about the change of nanoresonator decay rate. As has been noted it has smaller quality factor than quantum system one. Taking into account the fact that rate of nanoresonator dissipation consists of radiative and non-radiative losses we may write $\gamma_a^{rad} + \gamma_a^{nonrad} \gg \gamma_D$. When nanoresonator and quantum system approach to each other the rate of non-radiative decay in nanoresonator does not change because it depends on its material properties. At the same time the radiation rate γ_a^{rad} may significantly change due to changing of geometric structure of surrounding space, because the size of quantum system is comparable with the size of nanoresonator. Most significantly this change may occur for specific type of nanoresonators for which $\gamma_a^{rad} \gg \gamma_a^{nonrad}$, e.g. for "large" ($r \ge 30nm$) metallic nanoprticle or dielectric particle with whispering gallery mode. New rate of radiation $\tilde{\gamma}_a^{rad}$ may be much smaller than the relaxation rate of quantum dote $\gamma_D \gg \tilde{\gamma}_a^{rad} + \gamma_a^{nonrad}$. Thus nanoresonator becomes the oscillator with high quality factor in comparison with the quantum dot and it is possible to change the resonator decay rate (for this it is necessary to satisfy the inequality $\gamma_a^{rad} \gg \gamma_D \gg \tilde{\gamma}_a^{rad} + \gamma_a^{nonrad}$).

We now estimate how the rate of nanoresonator radiative losses may change in the presence of the quantum dot. Assume that there are several quantum dots where the main losses are nonradiative which around nanoresonator. We also assume that the size of nanoparticles and quantum dots is much smaller than the wavelength, $r \ll \lambda$. Then the absorption cross section of one quantum dot is $\sim \lambda^2$. We assume that the quantum dots surround nanoresonator, so we actually have "absorbing" cylinder around it with haracteristic height $\sim \lambda$ and size of base is $\sim r$. Nanoresonator can irradiate just through these bases. In the case of absence of quantum dots it radiated into the solid angle 4π , now it will radiate into the solid angle $\sim (r/\lambda)^2$. Since the radiation rate of nanoresonator is determined by integrating the Poynting vector over the surface through which it emits, we can estimate the change in the rate of radiation as $\tilde{\gamma}_a^{rad}/\gamma_a^{rad} \sim (r/\lambda)^2$. Since the characteristic size of the quantum dots and nanoresonator $\sim 50nm$, and wavelength $\sim 500nm$, we obtain the change in the rate of radiation by two orders.

This makes it possible to satisfy the inequality $\gamma_a^{rad} \gg \gamma_D \gg \tilde{\gamma}_a^{rad}$. For example, for semiconductor quantum dots $\gamma_D \sim 10^{11} s^{-1}$, characteristic radiative loss dielectric microsphere is $\gamma_a^{rad} \sim 10^{12} s^{-1}$, then $\tilde{\gamma}_a^{rad} \sim (r/\lambda)^2 \gamma_a^{rad} \sim 10^{10} s^{-1}$ and we have $\gamma_a^{rad} \gg \gamma_D \gg \tilde{\gamma}_a^{rad}$. These estimates show that the change in the Q factor of open resonator may be achievable in the experiment.

Now we return to the problem concerning the change of the open nanoresonator relaxation rate near quantum dot. The dynamics of their interaction is described by the system (4) - (7), but as an initial condition we assume the following. Let the atom at the initial time be in the ground state, and the cavity mode has a single quantum of energy (plasmon). We are interested in the decay rate of the resonator mode in this case. Dynamics as before is described by equations (4) - (7), but in this situation we choice the following initial condition $\langle \hat{a}^+ \hat{a} \rangle_{t=0} = 1$, $\langle D \rangle_{t=0} = -1$, $\langle \hat{\sigma}^+ a - \hat{a}^+ \hat{\sigma} \rangle_{t=0} = 0$. Also, we shall assume that at the initial time two-level system and cavity mode uncorrelated, so $\langle \hat{a}^+ \hat{\sigma}_z \hat{a} \rangle = \langle \hat{a}^+ \hat{a} \rangle \langle \hat{\sigma}_z \rangle = -1$. Then the dependence of number of plasmon on the time take the form:

$$\begin{aligned} \langle \hat{a}^{+} \hat{a} \rangle_{t} &= \frac{\exp(-\tau/2)}{\Gamma^{2}(1-16\Gamma^{2})} \left\{ \left(\Gamma^{2} + 8\alpha^{2}\Gamma^{2} - 16\Gamma^{4} \right) \cosh\left(\sqrt{1-16\Gamma^{2}}\tau/2\right) + \right. \\ &\left. + \left(2\Gamma^{2}\delta - \Gamma^{2} \right) \sqrt{1-16\Gamma^{2}} \sinh\left(\sqrt{1-16\Gamma^{2}}\tau/2\right) - 8\alpha^{2}\Gamma^{2} \right\}, \end{aligned}$$

where

$$\alpha = \Omega_R / \left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} + \gamma_D \right)$$

$$\begin{split} \Gamma = \sqrt{\frac{\Omega_R^2}{\left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} + \gamma_D\right)^2} + \frac{\left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad}\right)\gamma_D}{4\left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} + \gamma_D\right)^2}} \\ \delta = \gamma_D / \left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} + \gamma_D\right), \\ \tau = \left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} + \gamma_D\right)t. \end{split}$$

In the weak-coupling regime $|\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} - \gamma_D| \gg 4\Omega_R$ under condition $\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} \ll \gamma_D$ the time dependence of the plasmon number is following:

$$\left\langle \hat{a}^{+}\hat{a}\right\rangle_{t} = \exp\left(-\left(4\Omega_{R}^{2} + \left(\gamma_{a}^{nonrad} + \tilde{\gamma}_{a}^{rad}\right)\gamma_{D}\right)t/\left(\gamma_{a}^{nonrad} + \tilde{\gamma}_{a}^{rad} + \gamma_{D}\right)\right).$$
(23)

with the rate

$$\gamma_a^{tot} = \left(4\Omega_R^2 + \left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad}\right)\gamma_D^{nonrad}\right) / \left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} + \gamma_D^{nonrad}\right).$$
(24)

These expressions have the very important consequence. They are absolutely analogous to the equation (9) - (10) for population inversion damping. It means that we can introduce the Purcell factor for cavity mode. As for atom it describes the change of resonator damping, or quality factor, in the presence of two-level system.

The ratio of rates of cavity mode decay in the presence of a two-level system and without it in the limit $\gamma_a \gg 2\Omega_R \gg \gamma_D$ is described by the following equation

$$P_a = \frac{\left(4\Omega_R^2 + \left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad}\right)\gamma_D\right)}{\left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} + \gamma_D^{nonrad}\right)\left(\gamma_a^{nonrad} + \gamma_a^{rad}\right)},\tag{25}$$

and in the limit $\gamma_a \gg 2\Omega_R \gg \gamma_D$ we obtain

$$P_a = \frac{\left(4\Omega_R^2 + \left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad}\right)\gamma_D\right)}{\left(\gamma_a^{nonrad} + \tilde{\gamma}_a^{rad} + \gamma_D^{nonrad}\right)\left(\gamma_a^{nonrad} + \gamma_a^{rad}\right)}.$$
(26)

It should be noted that the decrease in the rate of radiation leads to a decrease in the intensity of radiation because of $\tilde{I}_{rad} \sim \tilde{\gamma}_a^{rad} \ll I_{rad} \sim \gamma_a^{rad}$. However, it is important that there is a significant narrowing of the line in $P_a \approx 4\Omega_R^2/\gamma_a^{rad}\gamma_D^{nonrad}$ times, that makes nanoresonator having larger quality factor, though with less radiative intensity.



Fig. 1 – (Color online) The dependence of the nanoresonator Purcell Factor P_a on the on the Rabi frequency Ω_R and nonradiative decay rate of the TLS γ_a^{nonrad} for fixed values of radiative and nonradiative nanoresonator decay rates, $\gamma_a^{rad} = 10^{13} s^{-1}$, $\gamma_a^{nonrad} = 10^{13} s^{-1}$. The values of Ω_R and γ_a^{nonrad} are chosen such that the condition $\gamma_D \gg 2\Omega_R \gg \gamma_a$ is satisfied

Thus, for the resonator we can apply the same arguments as for the two-level system. Changing the environment (in other words - the local density of states) leads to an effective change in the quality factor of the resonator. As shown previously for the two-level system, if heat losses are dominated in the cavity damping, the decay rate will be determined only by them. However, if the main part of the losses is associated with the radiation of the open resonator in the free space (for example, the resonator on the whispering gallery mode of dielectric sphere or plasmonic modes on large nanoparticles), the decay rate will be significantly changed. In this case, change of the channel of the dissipation of energy takes place and the main part of dissipation corresponds to the exciting of two-level system.

4. Discussion

We now discuss the physical meaning of obtained results. As shown in the previous section, quality factor of open nanoresonator changes significantly in the presence of a two-level system in the case when the main part of the energy dissipation corresponds to radiation. This effect is closely related to the Purcell effect for two-level atom, but in this case it tale place for the open resonator.

It is well know that the linewidth is determined by the quality factor of open resonator. Using the above calculations, we can estimate the characteristic change in the resonator linewidth as $4\Omega_R^2/\gamma_D\gamma_a^{rad}$. Depending on properties of the two-level system and the distance to it, this value can be greater or smaller than unity. Here, however, we need to take into account the following.

Expression $4\Omega_R^2/\gamma_D\gamma_a^{rad}$ tends to zero as the distance between the open nanoresonator and the two-level system goes to infinity. Of course, this is not valid. The fact is that everywhere we assume that nanoresonator interacts only with the two-level system by the near field. However, it also interacts with the modes of free space. When the two-level system is plased in the nanoresonator near field, this interaction is small compared to the near-field interaction between them, and equation (12) is valid. By increasing the distance between the two-level system and nanoresonator radiation to the modes of free space begins to dominate, and the rate of damping tends to γ_a^{rad} .

Thus, there are two competing processes: radiation into free space with the rate γ_a^{rad} and excitation of the two-level system with the finite lifetime and the damping associated with this process with the rate $4\Omega_R^2/\gamma_D$. As the distance between the open nanoresonator and two-level system will increase the rate of damping tends to γ_a^{rad} . The value $4\Omega_R^2/\gamma_D$ can be either greater or less than radiation into free space mode γ_a^{rad} .

It should be noted that linewidth of the nanolaser radiated field is very important for its operation. In the case of coupling with the dipole mode of a two-levelr system, an open cavity will radiate, but now with the rate $4\Omega_R^2/\gamma_D$. This enables to efficiently narrow linewidth of nanolaser operating in pulse mode.

5. Conclusions

The paper considers the Q factor of an open plasmonic resonator in the limit of small number of excited plasmons in it. It is shown that the Q factor is essentially determined by discrete nature of the energy excitations of the cavity. Practically, if the number of excited level in the resonator is of the order of unity it becomes a boson two-level system. Despite the difference statistics of quants in electronical TLS and in plasmonic cavity, it is possible to introduce a Purcell factor of the open plasmonic resonator cavity can also be d, showing the change of the resonator Q-factor due to changing of environment.

It is shown that in the case where the thermal cavity losses are small, the presence of QD may reduce the rate of decay of the excited mode resonator on several order. This is due to the fact that most of the energy stored in the cavity will go to the excitation of the QD. This phenomenon may be important, for example, to narrow nanolaser linewidth.

On the other hand, the rate of decay of the excited state of QD increases with rapproachment plasmonic nanoparticle to it. However, this may prove to be uncritical, as is now being discussed in the literature because the main radiative mechanism of nanolaser is nanoparticle luminescence.

The authors thank Yu. E. Lozovik for a helpful discussion.

Список литературы

- Surface-Enhanced Raman Scattering /Ed. by Kneipp K., Moskovits M., Kneipp H. Dordrecht: Springer, 2006.
- [2] Surface Enhanced Raman Spectroscopy: analytical, biophysical and life science applications /Ed. by Schlucker S. Weinheim: Wiley, 2011.
- [3] Pettinger B., Ren B., Picardi G., et al. // Physical review letters. 2004. V. 92. N. 9. P. 207412.
- [4] Bailo E., Deckert V. // Chemical Society Reviews. 2008. V. 37. N. 5. P. 921.
- [5] Deckert V., Zeisel D., Zenobi R., Vo-Dinh T. // Analytical Chemistry. 1998. V. 70. N. 13. P. 2646.
- [6] Heinzelmann H., Pohl D. W. // Applied Physics a-Materials Science & Processing. 1994. V. 59. N. 2. P. 89.

- [7] Keilmann F., Hillenbrand R. // Philosophical Transactions of the Royal Society of London Series a-Mathematical Physical and Engineering Sciences. 2004. V. 362. N. 787. P. 1817.
- [8] Koglin J., Fischer U. C., Fuchs H. // Physical Review B. 1997. V. 55. N. 12. P. 7977.
- [9] Webster S., Batchelder D. N., Smith D. A. // Applied Physics Letters. 1998. V. 72. N. 12. P. 1478.
- [10] Protsenko I. E., Uskov A. V., Zaimidoroga O. A., et al. // Phys. Rev. A. 2005. V. 71. N. 6. P. 063812.
- [11] Protsenko I. E. // Phys. Usp. 2012. V. 55. N. 1. P. 1040.
- [12] Berndt R., Gimzewski J. K., Johansson P. // Physical review letters. 1991. V. 67. N. 27. P. 3796.
- [13] Blackie E. J., Le Ru E. C., Etchegoin P. G. // Journal of the American Chemical Society. 2009. V. 131. N. 40. P. 14466.
- [14] Blackie E. J., Le Ru E. C., Meyer M., Etchegoin P. G. // J. Phys. Chem. 2007. V. 111. N. 3. P. 13794.
- [15] Lu Y.-J., Kim J., Chen H.-Y., et al. // Science. 2012. V. 337. N. 3. P. 6093.
- [16] Andrianov E. S., Pukhov A. A., Dorofeenko A. V., et al. // Phys. Rev. B. 2012. V. 85. N. 1. P. 035405.
- [17] Bergman D. J., Stockman M. I. // Physical review letters. 2003. V. 90. N. 2. P. 027402.
- [18] Noginov M. A., Zhu G., Belgrave A. M., et al. // Nature. 2009. V. 460. N. 7259. P. 1110.
- [19] Vinogradov A. P., Andrianov E. S., Pukhov A. A., et al. // Phys. Usp. 2012. V. 55. N. 3. P. 1046.
- [20] Andrianov E. S., Baranov D. G., Pukhov A. A., et al. // ArXiv cond-mat/1209.0422. 2012.
- [21] Pendry J. B., Maier S. A. // Physical review letters. 2011. V. 107. N. 12. P. 259703.
- [22] Stockman M. I. // Physical review letters. 2011. V. 106. N. 4. P. 156802.
- [23] Stockman M. I. // Physical review letters. 2011. V. 107. N. 5. P. 259704.
- [24] Wuestner S., Pusch A., Tsakmakidis K. L., et al. // Physical review letters. 2010. V. 105. N. 12. P. 127401.
- [25] Purcell E. M. // Physical Review. 1946. V. 69. N. 11-12. P. 681.
- [26] Oraevskii A. N. // Phys. Usp. 1994. V. 37. N. 4. P. 393.
- [27] Scully M. O., Zubairy M. S. Quantum Optics. Cambridge: Cambridge University Press, 1997.
- [28] Sachdev S. // Phys. Rev. A. 1984. V. 29. N. 5. P. 2627.

СУЖЕНИЕ ШИРИНЫ ЛИНИИ ПЛАЗМОННОГО НАНОЛАЗЕРА ПРИ УВЕЛИЧЕНИИ КОНСТАНТЫ СВЯЗИ ДВУХУРОВНЕВОЙ СИСТЕМЫ С ПЛАЗМОННОЙ НАНОЧАСТИЦЕЙ

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Аннотация

Исследовано излучение плазмонного нанолазера, состоящего из квантованного открытого резонатора и атома в сильно квантовом режиме, когда число возбужденных уровней в резонаторе порядка единицы. Ввиду сопоставимости размеров атома и резонатора следует учитывать фактор Парселла как для атома, так и для резонатора. Показано, что взаимодействие открытого нанорезонатора с атомом может уменьшить скорость излучения нанолазера и, следовательно, вызвать сужение ширины линии резонатора.

Ключевые слова: нанолазер, двухуровневая система, плазмонная наночастица, открытый резонатор, фактор Парселла