Dipole nano-laser

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Abstract. Theoretically predicted "dipole lasing", i.e., spontaneous excitation of coherent metal nano-particle dipole oscillations through interaction with a quantum-dot two-level system subject to population inversion is demonstrated. Equations for dipole lasing are the same as equations for ordinary laser, where the dipole momentum of nano-particle stands for the electromagnetic field cavity mode. Dipole lasing frequency corresponds to the localized plasmon resonance of the nano-particle. Dipole momentum of nano-particle leads to coherent dipole radiation. Optical cavity is not necessary, the size of the dipole laser can be smaller than the optical wavelength, i.e. it is dipole nano-laser. Threshold conditions and optical bistability in dipole nano-lasers are considered.

1. Introduction

Nano-particles can change dielectric and optical properties of the medium. Nano-particle of size $r_0 \ll \lambda/2\pi$ interacts with the electromagnetic field of wavelength $\lambda$ as a dipole; $N \gg 1$ dipoles uniformly distributed in space interact with the electromagnetic field the same way as a homogeneous medium [1]. Such nano-particles incorporated in a dielectric matrix form new "heterogeneous" medium (HM) with well-determined dielectric function $\varepsilon_{HM}$. HM dielectric function may be quite different from the dielectric functions of the nano-particle material and the material of the matrix. In case of metal nano-particles $\varepsilon_{HM}$ has a spike at $\lambda = \lambda_{LPR}$, this spike corresponds to Localized Plasmon Resonance (LPR). The value of $\lambda_{LPR}$ can be anywhere in the visible or near IR regions depending on the shape, the metal and the dielectric environment of the nano-particle [2].

HM has many practical applications, for example, as a filter of electromagnetic radiation [3] or for the increase of photovoltaic devices efficiency [4]. New interesting properties and applications of HM appear, when HM is "active", i.e. when HM has optical amplification at $\lambda \approx \lambda_{LPR}$. "Dipole lasing" discussed below and in [5] is novel physical phenomenon in HM with optical amplification, where it corresponds to a phase transition in the effective polarisability of nanoparticles [6]. Special properties of active HM were predicted in [7], similar phenomena have been investigated in [8, 9]. Recent experimental results point out to the possibility of the observation of dipole lasing in various systems. For example, sixfold increase in the Rayleigh scattering has been observed in a mixture of Rhodamine 6G dye and aggregates of silver nanoparticles pumped with a 532 nm Nd:YAG laser, when the pump increases a threshold [10]; quantum dot
luminescence enhanced by plasmons in the ordered planar structure of Ag nano-particles was observed in [11]; the emission of Si-based LEDs enhanced by plasmons of silver nano-particles was reported in [12]; the radiation of plasmonic laser antenna placed on the facet of a diode laser has been investigated in [13].

Here we present the theoretical analysis of the dipole lasing and related optical bistability as an extension of analysis of [5, 6]. In the next Section the equations for dipole laser are derived and threshold conditions are found, in Section 3 the optical bistability in dipole laser is considered. Results are summarized in Conclusion.

2. Equations and threshold conditions
A pair of quantum dot (q-dot) and the nano-particle is shown in Fig.1. Equations (1) - (3) describing optical properties of such system in the external monochromatic electromagnetic field of amplitude $E$ were derived in [5, 6]

$$
\dot{D} = 2i \left[ \Omega_{int} (a_+^* \sigma - \sigma^* a_0) + \frac{\mu_2}{\hbar} (\sigma E^* - E \sigma^*) \right] - (D - D_0)/\tau \tag{1}
$$

$$
\dot{\sigma} = (i \delta_0 - \Gamma) \sigma + i \left( \Omega_{int} a_0 - \frac{\mu_2 E}{\hbar} \right) D \tag{2}
$$

$$
\dot{a}_0 = (i \delta_0 - \Gamma_0) a_0 + i \left( \frac{\mu_0 E}{\hbar} - \Omega_{int} \sigma \right). \tag{3}
$$

Here $D = n - n_1$, $n$ and $n_1$ are populations of the energy states of q-dot, $D_0$ is the normalized pump rate, $a_0$ and $\sigma$ are normalized dipole momenta of the nano-particle and q-dot, respectively, $D$, $\sigma$, $a_0$ are quantum-mechanical operators the same as in the ordinary laser theory, $\mu_{0,2}$ are proportionality coefficients such that $d_0 = \mu_0 a_0$, $d_2 = \mu_2 \sigma$ are dipole momenta of the nano-particle and q-dot, respectively; we suppose $kr \ll 1$, $r$ is the distance between q-dot and the nano-particle, $k = 2\pi/\lambda_{LP R}$; $\Omega_{int} \approx \xi \mu_0 \mu_2/(\hbar r^3)$ is the rate of the dipole-dipole interaction between the nano-particle and q-dot, $\xi = 2$ for the case shown in Fig.1, $\delta_0 = \omega - \omega_{LP R}$, $\delta_2 = \omega - \omega_{LP R}$, $\omega_{LP R}$ and $\omega_2$ are frequencies of the electric field, LPR and the q-dot transition, respectively; $\Gamma$, $\Gamma_0$ and $1/\tau$ are relaxation rates. By setting $\Omega_{int} = 0$ in Eqs.(1) - (3) (which corresponds to large distance between particles) one can show that $\mu_2^2 = \alpha_0 r \Gamma_0$, where $\alpha_0$ is the resonant (i.e. at $\delta_0 = 0$) polarisability of the nano-particle ($i = 0$) and the q-dot ($i = 2$), so that $\Omega_{int}^2 = 4\alpha_0 \alpha_2 \Gamma_0 \Gamma_2 / r^6$. One can find the stationary solution of Eqs.(1) - (3), calculate

\[\text{Figure 1. Nano-particles 1 on the dielectric layer 3 near the surface of the semiconductor medium 2 with q-dots (triangles) with the population inversion. Single dipole nano-laser is outlined by the dashed contour. Large arrows show directions of generated dipole momenta for spherical or ellipsoidal nano-particle corresponding the minimum value of the threshold.}\]

the polarisability $\alpha_{tot} = (\mu_0 a_0 + \mu_2 \sigma)/E$ of the nano-particle and q-dot and find the divergence $\alpha_{tot} \to \infty$ at the pump rate $D_0 = D_{th} = r^6/(4|\alpha_0||\alpha_2|)$, where $\alpha_i = \alpha_{ir}/(\delta_i/\Gamma_i - 1)$, $i = 0, 2$ is
the polarisability of the isolated nano-particle and q-dot. The divergence in $\alpha_{\text{tot}}$ means dipole lasing, i.e. spontaneous appearance of dipole momenta $d_0 = \mu_0 a_0 E \neq 0$, $d_2 = \mu_2 \sigma E \neq 0$ without external coherent field: $E = 0$ [6]. Eqs. (1) - (3) are the same as equations for ordinary laser, but the nano-particle dipole momentum (operator $a_0$) replaces there the cavity mode; this is why the system in Fig.1 is named as "dipole" laser. Dipole laser emits electromagnetic field as a dipole, it does not need optical cavity, so that its size may be smaller than the optical wavelength, i.e. it is dipole nano laser (DNL). Eqs. (1) - (3) are most simple laser equations, they demonstrate the principle idea of dipole laser. Eqs. (1) - (3) can be derived from three-level laser scheme, when the q-dot semiconductor environment plays the role of the third level pumped by injected current. The relaxation of the electron from the environment to the q-dot upper level assumed very fast, which lets us to replace three level scheme by two level scheme with the pump directly from the low to the upper level. Generalization of Eqs. (1) - (3) to more complex schemes, as the four level scheme, is straightforward as it is done in the conventional laser theory.

The requirement $D_0 < 1$ leads to the necessary condition for dipole lasing, that is small distance between the nano-particle and q-dot

$$r < r_{cr} = (4|\alpha_0||\alpha_2|)^{1/6}. \quad (4)$$

One can easily provide almost complete population inversion $D_0 \approx 1$ in q-dot, thus the most important condition for the dipole lasing is Eq. (4). The resonant (i.e. at $\delta_2 = 0$) polarisability of q-dot is $\alpha_2 = \frac{3}{4} \left( \frac{\lambda_{\text{LPR}}}{2\pi} \right)^2 \frac{\Gamma_{sp}}{\Gamma_2}$, where $\Gamma_{sp}$ is the spontaneous emission rate of q-dot transition (see, for example [14]). One has to provide LPR in the near IR range typical for q-dot resonant transitions. For that one has to take a needle-like [3] or spherical core and shell nano-particle. Polarisability of the core-shell nano-particle is [13]

$$\alpha_0 = r_0^3 \xi_{cs}, \quad \xi_{cs} = \frac{(\varepsilon_{b1} - \varepsilon_m)(\varepsilon_{b2} + 2\varepsilon_{b1}) + \beta(\varepsilon_{m} + 2\varepsilon_{b1})(\varepsilon_{b2} - \varepsilon_{b1})}{(\varepsilon_{b1} + 2\varepsilon_m)(\varepsilon_{b2} + 2\varepsilon_{b1}) + 2\beta(\varepsilon_{b1} - \varepsilon_m)(\varepsilon_{b2} - \varepsilon_{b1})}. \quad (5)$$

Here $\varepsilon_{b1}, \varepsilon_{b2}$ are the shell and the core dielectric functions, respectively, $\beta = \varepsilon_1/r_0$, $r_1$ is the core radius, $r_0$ is the radius of the nano-particle.

Fig.2 shows the maximum distance $r_{cr}(r_0)$ between q-dot and the nano-particle, when the nano-particle is made of the polymer core with $\varepsilon_{b2} = (1.55)^2$ and silver shell, such that $\beta = 0.6$. The nano-particle is in Si matrix, $\lambda_{\text{LPR}} = 1.13 \mu m$. One can see from Fig.2 that the necessary condition (4) for dipole lasing can be satisfied at low temperatures when $\Gamma_{sp}/\Gamma_2 > 10^{-2}$. In principle, one can make many such pairs on the surface of the semiconductor medium with q-dots [11] and provide collective coherent emission from many DNL. It is necessary to keep the distance $r < r_{cr}$ in each pair q-dot and nano-particle, though the distance between different pairs may be arbitrary.

3. Optical bistability

It is shown in [6] that DNL in the external field has optical bistability. For example, there are three different values of the DNL radiation rate $\gamma_{\text{rad}}(I)$ (see the curve 4 in Fig.3 of [6]) for the same value of the external field normalized intensity $I$. Here we find necessary conditions for the bistability in DNL in the catastrophe theory approach [16] and show that the optical bistability in the system shown in Fig.1 is possible also at the absorption and at room temperatures.

The necessary condition for the bistability in DNL at external field is when the solution of the equation

$$d\gamma_{\text{rad}}(I)/dI = \infty, \quad (6)$$

has physical sense, i.e. for this solution $I > 0, |D_0| < 1, |D| < 1, D < D_0$ and the distance $r$ between particles $r > r_0 + r_2$, where $r_0, r_2$ are the size of the nano-particle and q-dot, respectively.
Supposing that smooth function $I(\gamma_{rad})$ is inverted to the function $\gamma_{rad}(I)$, we can write the necessary condition for the bistability as

$$dI/d\gamma_{rad} = 0. \quad (7)$$

The function $\gamma_{rad}(I)$ in [6] is determined in the non-explicit (parametric) form, i.e. through $I(D)$ and $\gamma_{rad}(D)$ where $D$ is a parameter (see Eqs. (11) and (12) of [6], respectively). Thus one can write $\gamma_{rad}(I) = \gamma_{rad}(D(I))$, which means that Eq.(7) is equivalent to

$$\frac{\partial I}{\partial D} \frac{\partial D}{\partial \gamma_{rad}} = 0. \quad (8)$$

According with Eq.(12) of [6] $\frac{\partial \gamma_{rad}}{\partial D} \neq \infty$ so that the necessary condition for the bistability is

$$dI(D)/dD = 0. \quad (9)$$

The sufficient condition for the bistability in DNL corresponds to the appearance of the inflection point on the curve $I(D)$. Such points called also as bifurcation points in catastrophe theory. One finds the inflection (or the bifurcation) point on the curve $I(D)$ when

$$d^2 I(D)/dD^2 = 0 \quad (10)$$

is satisfied together with condition (9). Let us explain why Eqs.(9) and (10) give sufficient condition for the bistability. Suppose that some $I$, $D$, $D_0$, $r$ and other parameters are such that

**Figure 2.** The minimum distance $r_{cp}$ between borders of the nano-particle and q-dot as a function of radius $r_0$ of the core-shell silver nano-particle in Si at various linewidths $\Gamma_2$ of the q-dot transition. $\Gamma_{sp}/\Gamma_2 = 1$ (curve 1); 0.1 (2); 0.01 (3). The q-dot radius is 5 nm.
Eqs. (9) and (10) are satisfied, so that the curve \( I(D) \) has an inflection point, as the curve 1 shown on the insert in Fig.4. Let us slightly change some parameter (for example, \( D_0 \)), then the curve \( I(D) \) is deformed such that three different values of \( D \) for single \( I \) appear in some region of \( I \), see the curve 2 on the insert in Fig.4. Thus the inflection point marks the border in the parameter space separating the region with single \( D(I) \) (curve 1 on the insert in Fig.4) and the bistability region with three \( D \) for the same \( I \) (curve 2 on the insert in Fig.4).

Let us now determine sufficient condition for the bistability in DNL, when Eqs.(9) and (10) have common solution. We came back to equations (1) – (3) for DNL in the external field, find the stationary solution and obtain [6]

\[
I(D) = \left| \frac{1 + D \xi^2 \alpha_2 \alpha_0 / r^6}{1 - \xi \alpha_0 / r^3} \right|^2 \left( \frac{D_0}{D} - 1 \right),
\]

where \( \alpha_{0,2} \) are polarizabilities of the nano-particle (the index 0) and q-dot (the index 2), the value \( 1 \leq |\xi| \leq 2 \) depends on mutual orientation of q-dot and the nano-particle. Introducing new variables \( F(D) = I(D) \left| 1 - \xi \alpha_0 / r^3 \right|^2, \ x = D / D_0 \) and parameters \((1/D_*) e^{i \varphi} = \xi \alpha_0 \alpha_2 / r^6, \ d_0 = D_0 / D_* \) we write instead of Eq.(11)

\[\text{Figure 3. Regions of bistability in the absorption of HM consisted of pairs of silver ellipsoidal nano-particles and q-dots in Si matrix for } r = 9 \text{ nm and the temperature 100 K (curve 1) and 300 K (curve 2)}\]

\[
F(x) = \left[ 1 + 2d_0 \cos(\varphi) x + x^2 d_0^2 \right] (1/x - 1),
\]

and \( dF/dx = 0 \) instead of Eq.(9), which is equivalent to

\[
x^3 - \theta x^2 + 1/2 d_0^2 = 0,
\]
where \( \theta = (1/2)[1 - 2 \cos(\varphi)/d_0] \). We have \(3x^2 - 2\theta x = 0\) instead of Eq.(10), which gives two inflection points \(x_{inf} = 0\) and
\[
x_{inf} = \frac{1}{3}\left[1 - \frac{2 \cos(\varphi)}{d_0}\right].
\] (14)

The solution of Eq.(13) at \(x = x_{inf} = 0\) exists only at \(D_s = 0\), i.e. when \(r = 0\), which can’t be, so that there is only one inflection point given by Eq.(14). By inserting \(x_{inf}\) from Eq.(14) into Eq.(13) we obtain \(\theta^2 = 27/(8d_0^4)\), which can be resolved with the respect to \(\cos(\varphi)\)
\[
\cos(\varphi) = \frac{1}{2}\left(d_0 - 3d_0^{1/3}\right).
\] (15)

By inserting Eq.(15) into Eq.(14) we find \(x_{inf} = d_0^{-2/3}\). Because \(|\cos(\varphi)| \leq 1\), Eq.(15) determines the region of \(d_0\)
\[
|d_0 - 3d_0^{1/3}| \leq 2,
\] (16)
where one can find inflection points on the curve \(F(x)\). One can see that Eq.(16) is true for \(|d_0| < 8\). Because of the dimensionless intensity of the external field \(F(x) > 0\) at \(0 < x < 1\) and \(x_{inf} < 1\) at \(|d_0| > 1\), the region where inflection points exist is restricted by
\[
1 \leq |d_0| \leq 8,
\] (17)
and
\[
D_s \equiv r^6/(\xi|\alpha_0\alpha_2|) = 1
\] (18)
determines the maximum distance \(r\) between q-dot and the nano-particle when the bistability is possible. Note that, in general, \(D_s \neq D_{th}\) which is the threshold of dipole lasing given by Eq.(18) at \(\delta_0/\Gamma_0 = -\delta_2/\Gamma_2\). The maximum value of \(r\) necessary for the bistability corresponds to the exact resonance \(\delta_0 = \delta_2 = 0\), so that the bistability is not possible at \(r\) larger than the minimum \(r\) necessary for dipole lasing.

An interesting result is that the bistability is possible at \(D_0 < 0\) and \(D < 0\), i.e. at the absorption. For example, Fig.3 shows the bistability regions in the parameter space \((J,D_0)\), where \(J\) is the intensity of the external field. Fig.3 is for silver ellipsoidal nano-particle with the semi-axes \(a = 6.5\) nm and \(b = 3.25\) nm (for such silver nano-particle in \(Si\) \(\lambda_{LP} = 1.3\) \(\mu m\)) and q-dots placed in \(Si\) matrix, when the distance between the nano-particle and q-dot is 9 nm. One can see that the bistability regions (they are inside \(A\)-shaped curves in Fig.3) exist at the absorption \(-1 < D_0 < 0\) and at room temperatures. Fig.4 shows the absorption coefficient of the heterogeneous medium consisted of pairs of q-dots and the ellipsoidal nano-particles in \(Si\) with the density of pairs \(10^{14}\) \(cm^{-3}\). In order to obtain curves in Fig.4 we calculate the polarization \(\alpha_{tot}\) of single pair q-dot and nano-particle
\[
\alpha_{tot} = \left|\frac{\alpha_0 - \alpha_2 + \xi(\alpha_0\alpha_2)/r^3(D + 1)}{1 - \xi\alpha_0/r^3}\right|^2,
\] (19)
see Eq.(12) of [5], find \(\varepsilon_{HM}\) from the Maxwell-Garnett formula [17]
\[
\frac{\varepsilon_{HM} - \varepsilon_m}{\varepsilon_{HM} + 2\varepsilon_m} = \frac{4\pi}{3}N_0\alpha_{tot}.
\]
with \(\varepsilon_m = (3.4)^2\) and calculate absorption coefficient \(\kappa_{HM} = (2\pi/\lambda)[(\varepsilon_{HM} - \varepsilon_{HM}^{*})/2]^{1/2}\) of HM. The appearance of the bistability can be interpreted as a first order phase transition in the resonant HM with metal nano-particles and nonlinear absorption.
Figure 4. Bistability in the absorption coefficient of HM consisted of $N_0 = 10^{14}$ cm$^{-3}$ of pairs of q-dot and silver ellipsoidal nano-particles in Si matrix at room temperature, when the distance between q-dot and the nano-particle is 9 nm. Insert explains how many stationary solutions appear near the inflection point on the curve $I(D)$ (see the text).

4. Conclusion

Metal nano-particles let us to construct heterogeneous optical media (HM) with pre-determined localized plasmon resonances (LPR) in the dielectric function. Spontaneous appearance of macroscopic polarization, or "dipole lasing", is possible in HM at LPR with nonlinear optical amplification. Such amplification can be provided, for example, by quantum dots with the population inversion. Single pair of a nano-particle and q-dot is a dipole nano-laser, where coherent oscillations of the electron density appear at certain threshold conditions. Coherent oscillations of dipole momenta of particles lead to coherent dipole radiation. Such dipole laser does not need optical cavity, so its size may be smaller than optical wavelength, i.e. it is dipole nano laser (DNL). In practice DNL can be realized, for example, by placing nano-particles on the surface of semiconductor media with optical amplification. It is preferable to provide a "near order" in such structure, for example, by organizing nano-particles and q-dots in pairs with certain distance between particles in each pair. Estimations show, that the threshold pump current density in DNL is about 1 A/cm$^2$, the DNL output power is $\sim 150$ kW/cm$^3$, the quantum efficiency is $\sim 40\%$. Because of the low quality factor of LPR, dipole lasing can be observed, most probably, at low temperatures of few tens of K. The metal nano-particle and q-dot coupled through the dipole-dipole interaction at LPR has optical bistability in the external field at the absorption or amplification even at room temperatures.

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