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Theory of the dipole nanolaser

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1. Introduction

This paper is concerned with the theoretical investigation into the coherent generation of the dipole moment of a metallic nanoparticle, when a localized plasmon resonance is excited in it with the use of a generator (an atom, a quantum dot, etc.) in which an inverse electron state population is realized. The generator and the nanoparticle experience a near-field interaction. This nano-dimensional system-a 'dipole' nanolaser (DNL)-emits a coherent electromagnetic field when threshold conditions are fulfilled. We derive basic and simplified DNL equations, discuss the threshold lasing conditions and the DNL features associated with incoherent dipole moment generation, and take a look at several DNL-based facilities (broadband optical modulators, highly efficient light-emitting devices), as well as possible DNL designs and further lines of their theoretical investigation.

Research into the resonance interaction of electromagnetic radiation with metallic particles have a long-standing history [1–3]. Today, these studies are on the rise, and a new field of physics — nanoplasmonics [4] — has made its appearance, which is related to the development of nanotechnologies and the emergence of new practical tasks: to control light in optoelectronics [5, 6], enhance the efficiency of solar elements using metallic nanoparticles [7], etc. Metallic nanoparticles possessing localized plasmon resonances (LPRs) may be employed as antennas in the optical and near-infrared regions [8], including the production of nano-dimensional lasers. The method of coherent radiation generation and the dipole nanolaser based on this were proposed in patent [9]; the corresponding theory was elaborated in Refs [10-17] and applied, for instance, in Refs [18, 19]. Independently of and practically simultaneously with Ref. [9], the authors of patent [20] came up with a method of generating surface plasmons and a device for surface plasmon amplification by stimulated emission of radiation (spaser), which is close to the DNL. The theory of spacers was elaborated in Ref. [21] and in other work. Also investigated was the possibility of compensating losses in active media with metallic nanoparticles [22]; this loss compensation takes place both in DNLs and in spasers. Several experiments have been carried out with devices close to the DNL, which were operated both below [23, 24] and above [25, 26] the lasing threshold; the complete bibliography of DNL and spaser research is hard to expound on in this short report.

The aim of this paper is to briefly outline the theory of DNLs and to consider some of its new features. The DNL

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Uspekhi Fizicheskikh Nauk **182** (10) 1116–1122 (2012) DOI: 10.3367/UFNr.0182.201210i.1116 Translated by E N Ragozin; edited by A Radzig theory is based on the well-known equations of a single-mode laser [27], in which the electromagnetic field mode is replaced with the dipole moment arising from the resonance oscillations of the nanoparticle electron density. The quantum of linear electron density oscillations is a boson, like the quantum of an electromagnetic field (EMF), and coherent (laser) generation of electron density oscillations is therefore possible, similarly to coherent generation of EMF bosons. The feasibility of laser generation of not only EMF quanta, but also other bosons, for instance, Bose-Einstein condensates of atoms in traps [28], was emphasized earlier. Basic DNL equations substantiate the possibility of coherent generation not only of the dipole moment and radiation but of other laser effects in DNL-like systems as well. The elaboration of the basic equations results in progressively more realistic models of the DNL and its related devices. In doing so, it is possible to employ the well-known approaches of laser theory, for instance, in the description of spontaneous emission into a mode [29, 30], as well as of classical electrodynamics, for instance, in the description of DNL modes [3]. Those effects which are of little significance in 'classical' laser theory quite often turn out to be highly important for the DNL like, for instance, nonthreshold lasing [30] or local field effects [31]. Elaborating the basic DNL equations leads to models different from their analogs in laser theory; in particular, the equations for the DNL in an external field with the inclusion of spontaneous emission into a mode [17] are significantly different from the equations for an ordinary laser with external signal injection [27]. The DNL theory is highly instructive, because it combines, in a natural way, the classical effects of the nonlinear dynamics of oscillatory systems (threshold effects, bistability, etc.) and the special features of quantum electrodynamics (spontaneous emission, quantum noise, single-photon emission). This all makes the DNL an interesting subject of basic research, but its practical applications are equally interesting

The paper outline is as follows. In Section 2, we offer the derivation of DNL equations, which is more general than in Ref. [10], without invoking the quasistatic approximation and the assumption that the nanoparticle dimensions are negligibly small. In Section 3, we simplify the basic equations: they are brought to the form of Ref. [10] convenient for applications; validity conditions of the equations are discussed and many-atom DNL equations are considered. Sections 4 and 5 are concerned with the DNL oscillation conditions and point to the possibility of bifurcations in lasing, when the direction of dipoles changes in a step-wise manner; also included is the incoherent (spontaneous) generation of the dipole moment of a few-atom DNL. In summary, we discuss the prospects of DNL theory development.

2. Basic equations of the dipole nanolaser

A simple DNL design comprises a metallic nanoparticle and a nearby emitting two-level system (atom, molecule, quantum dot, etc.) in which the population inversion of its states is effected with the aid of an incoherent external pump: a broadband light pulse, injection current, and so forth. (Fig. 1a). The localized plasmon resonance frequency ω_{LPR} of the metallic nanoparticle is close to the transition frequency ω_2 of the two-level system; the particles experience resonance interaction via EMF.

The equations for a two-level system [32] in an external resonance field $\mathcal{E} = \mathbf{e}[E(t) \exp(-i\omega t) + E^+(t) \exp(i\omega t)]$ are

Figure 1. DNL diagrams: (a) metallic nanoparticle 1 is separated from the two-level system — quantum dot 3 — by a dielectric sublayer 2; (b) 'coreshell' type DNL.

well known, where **e** is a unit vector, and the amplitude E(t) varies more slowly than exp $(-i\omega t)$. In the approximation of slowly varying amplitudes, one has

$$\dot{D} = \frac{2i\mu e}{\hbar} (E\sigma^{+} - E^{+}\sigma) - \tau^{-1}(D - D_{0}), \qquad (1)$$

$$\dot{\sigma} = (\mathrm{i}\delta_2 - \Gamma_2)\sigma - \mathrm{i}\frac{\mu \mathbf{e}}{\hbar} ED, \qquad (2)$$

where *D* is the population inversion; $\boldsymbol{\mu}$ is the real dipole matrix element; τ^{-1} and Γ_2 are the relaxation rates of the population of the upper level in the two-level system and of its dipole moment, respectively; D_0 is the pump-induced equilibrium value of population inversion in the absence of the field, and $\delta_2 = \omega - \omega_2 \ll \omega_2$; the dipole moment of the two-level system $\hat{\mathbf{d}}_2 = \boldsymbol{\mu}[\sigma \exp(i\omega t) + \sigma^+ \exp(-i\omega t)]$, and its magnetic moment is assumed to be zero; the resonance frequency of the transition between the states is $\omega_2 \approx \omega_{\text{LPR}}$.

We replace σ in Eqn (2) by Bose operator a, $[a^+, a] = 1$, and at D = -1 arrive at the equation of motion for the dipole moment $\hat{\mathbf{d}}_0 = \boldsymbol{\mu}_0[a \exp(i\omega t) + a^+ \exp(-i\omega t)]$ of the nanoparticle, i.e. the linear dipole (harmonic oscillator) in the resonance EMF:

$$\dot{a} = (\mathrm{i}\delta - \Gamma_{\mathrm{LPR}})a + \mathrm{i}\,\frac{\mu_0 \mathbf{e}}{\hbar}\,E\,,\tag{3}$$

where $\delta = \omega - \omega_{LPR} \ll \omega_{LPR}$, Γ_{LPR} is the LPR linewidth, and *a* may be termed the plasmon annihilation operator.

The two-level system is treated as a point dipole. The finite nanoparticle dimensions should be taken into consideration: the two-level system may be located closely to the nanoparticle, so that the field of the system changes markedly over the domain occupied by the nanoparticle. As a rule, the nanoparticle dimensions are smaller than the EMF wavelength, and the quasistatic approximation therefore applies inside the nanoparticle, and the polarization (the dipole moment of a unit volume) is the same throughout the nanoparticle [4]. Let us introduce the Bose operator $p(\mathbf{r})$ of polarization amplitude, $a = \int_V p(\mathbf{r}) dV$, where V is the nanoparticle volume. The commutation relations $[p(\mathbf{r}), p^+(\mathbf{r}')] = V\delta(\mathbf{r} - \mathbf{r}')$ ensure that $[a, a^+] = 1$ for the Bose operator a; \mathbf{r} and $\mathbf{r'}$ are the radius vectors of the points inside the nanoparticle. The equation of motion for $p(\mathbf{r})$ has the form

$$\dot{p}(\mathbf{r}) = (\mathrm{i}\delta - \Gamma_{\mathrm{LPR}}) p(\mathbf{r}) + \mathrm{i} \, \frac{\mu_0 \mathbf{e}}{V\hbar} \, E(\mathbf{r}) \,.$$

By integrating both sides of this equation over the nanoparticle volume, we obtain

$$\dot{a} = (\mathrm{i}\delta - \Gamma_{\mathrm{LPR}})a + \mathrm{i}\frac{\mu_0}{\hbar}\bar{E}_{\mu}, \qquad (4)$$
$$\bar{E}_{\mu}(r) = V^{-1} \int_{V} \mathbf{e}_{\mu} \mathbf{e}(\mathbf{r}') E(\mathbf{r}') \,\mathrm{d}^3 r',$$

where \bar{E}_{μ} is the nanoparticle volume-averaged projection of the external field amplitude onto the polarization mode under excitation (in the present case, onto the direction of the nanoparticle's dipole moment), \mathbf{e}_{μ} is the unit vector aligned with the nanoparticle's dipole moment, and *r* is the nanoparticle center distance from the two-level system. For a spherical nanoparticle, $\bar{E}_{\mu}(r) = E(\mathbf{r})$ in the quasistatic approximation, which can be directly verified.

Now, it is possible to describe the interaction, via an alternating electric field, between the two-level system and the metallic nanoparticle, which are located in a uniform transparent medium with a refractive index n_2 and separated by some distance. The electric field induced by the particle in the seat of the two-level system will be denoted by the subscript 02, while the field produced by the two-level system in the seat of the nanoparticle will be denoted by the subscript 20. We use Eqns (1)–(3) to arrive at the system of equations of motion for both dipoles — the particle and the two-level system — with the inclusion of their dipole–dipole interaction:

$$\dot{D} = 2i \,\frac{\mu \mathbf{e}_{02}}{\hbar} (\sigma^+ E_{02} - E_{02}^+ \sigma) - \tau^{-1} (D - D_0) \,, \tag{5}$$

$$\dot{\sigma} = (\mathrm{i}\delta_2 - \Gamma_2)\sigma - \mathrm{i}\,\frac{\mu \mathbf{e}_{02}}{\hbar}\,E_{02}D\,,\tag{6}$$

$$\dot{a} = (\mathrm{i}\delta - \Gamma_{\mathrm{LPR}})a + \mathrm{i}\,\frac{\langle \boldsymbol{\mu}_0 \mathbf{e}_{20} E_{20} \rangle_V}{\hbar}\,,\tag{7}$$

where $\langle \ldots \rangle_V$ signifies averaging over the nanoparticle volume.

By using the expression for the Fourier component of the dipole radiation [33], the approximation of slowly varying amplitudes, and the spectral decomposition of $E_{02}(t)$ and $E_{20}(t)$ into Fourier integrals, it is possible to show that the electric field amplitudes entering into equations (5)–(7) are written down as

$$\boldsymbol{\mu} \mathbf{e}_{02} E_{02}(t) = -\frac{\mu_0 \mu}{r^3} \, \xi(k_2 r) \sigma \left(t - \frac{n_2 r}{c_0} \right),$$

$$\boldsymbol{\mu}_0 \mathbf{e}_{20} E_{20}(t) = -\frac{\mu_0 \mu}{r^3} \, \xi^*(k_2 r) a \left(t - \frac{n_2 r}{c_0} \right),$$

$$\xi(\rho) = \exp\left(i\rho\right) \left[(1 - \rho^2 - i\rho) + \cos^2\theta_r(\rho^2 + 3i\rho - 3) \right],$$
(8)

where $\rho = k_2 r$, *r* is the observation point distance from the dipole, $k_2 = n_2 \omega/c_0$, **n** is a unit vector directed from the dipole to the point of observation, and θ_r is the angle between **n** and parallel vectors $\mathbf{d}_{0,2}$; this angle will be determined below. In the second of expressions (8), ξ^* appears, while ξ enters into the first one, because the fields from particle 0 to particle 2 and vice versa propagate in opposite directions. On the right sides of expressions (8) there are dipole polarization amplitudes delayed by a time $n_2 r/c_0$ taken for the EMF to travel the interparticle distance.

Equations (5)–(8) constitute the DNL basic equations. Their variables are the population inversion D and the dipole moments σ and a. Electromagnetic fields do not enter into the equations, and the fields outside of the DNL may be determined from expressions (8) and the corresponding expressions for the magnetic fields of the dipoles. In the DNL, the place of a field mode is occupied by nanoparticle polarization (dipole moment) mode a, which gave rise to the name 'dipole' nanolaser. Instead of the polarization of the nanoparticle, it is possible to consider the field inside it; in the quasistatic approximation and under the excitation of a 'dipole' mode, this field differs from the polarization only by a factor standing for the electric susceptibility of the nanoparticle material. In a similar way, it is possible to derive equations when another electron density oscillation mode, for instance, the quadrupole one with a different LPR frequency, rather than the dipole mode, is excited in the nanoparticle. The oscillation modes for the field (or polarization) inside and outside spherical particles are exactly known [3]: they may be applied to analyzing the DNL with large spherical particles, when the quasistatic approximation is invalid inside them. For small nonspherical nanoparticles, it is possible to use the quasistatic approximation when describing the particle polarizations (the fields inside the particles) and multipole expansions to describe the fields outside the particles [33].

Equations (5)–(8) are operator equations, and their variables are fluctuating quantities with intrinsic quantum fluctuations and those caused by dissipation. One way to solve such equations involves the employment of spectral decompositions and the addition of Langevin forces, as was done in Ref. [30]. To analyze the DLN, we replace below equations (5)–(8) with equations for the averages and 'uncouple' correlations (an analog of the moments method in statistical theory): higher-order moments, i.e., the averages of the products of fluctuating quantities, are replaced by lower-order moments — by the products of the averages.

3. Simplified equations, their validity conditions, and conditions for dipole nanolaser oscillation

Let us assume that the interparticle distance is short, so that the EMF delay time $\tau_r = n_2 r/c_0$ is short in comparison with the characteristic time of change of variables, and in Eqns (8) we can write

$$\sigma\left(t - \frac{n_2 r}{c_0}\right) \approx \sigma(t) - \tau_r \frac{\mathrm{d}\sigma}{\mathrm{d}t}, \qquad a\left(t - \frac{n_2 r}{c_0}\right) \approx a(t) - \tau_r \frac{\mathrm{d}a}{\mathrm{d}t}.$$
(9)

For sufficiently short *r*, when $\tau_r d\sigma/dt \ll \sigma(t)$ and $\tau_r da/dt \ll a(t)$, the derivatives on the right-hand sides of expressions (9) can be neglected, and then the system of equations (5)–(7) assumes the form

$$\dot{D} = 2\mathrm{i}(\Omega_{\mathrm{int}}^* a^+ \sigma - \Omega_{\mathrm{int}} \sigma^+ a) - \tau^{-1} (D - D_0), \qquad (10)$$

$$\dot{\sigma} = (\mathrm{i}\delta_2 - \Gamma_2)\sigma + \mathrm{i}\Omega_{\mathrm{int}}aD\,,\tag{11}$$

$$\dot{a} = (\mathrm{i}\delta - \Gamma_{\mathrm{LPR}})a - \mathrm{i}\bar{\Omega}_{\mathrm{int}}^*\sigma\,,\tag{12}$$

where the coupling constant

$$\Omega_{\rm int} = \xi(k_2 r) \,\frac{\mu_0 \,\mu}{\hbar r^3} \,, \qquad \bar{\Omega}_{\rm int}^* = \frac{1}{V} \int_V \Omega_{\rm int}^* \,\mathrm{d}V \,, \tag{13}$$

 $\Omega_{\text{int}} = \Omega_{\text{int}}(r)$, $\overline{\Omega}_{\text{int}}^* = \overline{\Omega}_{\text{int}}^*(r)$, and *r* is the two-level system distance from the nanoparticle center. For a spherical nanoparticle of radius *a*, in the quasistatic approximation,

when $k_2a < k_2r \ll 1$, it turns out that $\bar{\Omega}_{int}^* = \Omega_{int}^*$. On the other hand, beyond the quasistatic approximation and for small particles, when $k_2a \ll 1$ and $k_2r \ge 1$, we may put $\bar{\Omega}_{int} = \Omega_{int}$, correct to at least $\sim a/r \ll 1$. Therefore, for small spherical nanoparticles if nothing else, it may be assumed that the condition $\bar{\Omega}_{int} = \Omega_{int}$ is always fulfilled with a high accuracy.

DNL equations (10)–(13) coincide with single-mode laser equations, for instance (on changing the notation) with Eqns (3.2) from Ref. [27]. This signifies that it is possible to directly take advantage of several results of laser theory in the DNL analysis, including the possibility of the coherent generation of a nanoparticle dipole moment by analogy with the coherent EMF mode generation in a conventional laser. As in the latter, the DNL may have a lasing threshold, although DNLs quite often turn out to be 'nonthreshold' (see below), unlike ordinary lasers which are nonthreshold only under special conditions [30].

Let us estimate when it is possible to neglect retardation in Eqns (8). The DNL operates (reaches a stationary mode, etc.) with a characteristic rate $\sim \Gamma_{\rm LPR}$, i.e., $\dot{\sigma}/\sigma \sim \dot{a}/a \sim \Gamma_{\rm LPR}$. The LPR quality factor $Q = \omega_{\rm LPR}/(2\Gamma_{\rm LPR})$. Considering these relations, we find the smallness condition for the delay time $\tau_{\rm r} = n_2 r/c_0$:

$$\frac{n_2 r}{c_0} \Gamma_{\rm LPR} = \frac{n_2 r}{c_0} \frac{\omega_{\rm LPR}}{2Q} = \frac{k_2 r}{2Q} \ll 1 \text{ or } r \ll r_{\rm cr} = \frac{2Q}{k_2} .$$
(14)

Figure 2 depicts the dependence of Q on the radius a of a spherical gold nanoparticle in water, $n_2 = 1.33$. For the peak value of Q = 33.8 for a = 19 nm and $\lambda_{LPR} \equiv \omega_{LPR}/c_0 = 560$ nm, we obtain the validity condition for equations (10)–(12): $r \ll r_{cr} = 4.53$ µm. Therefore, when the atom is located at a distance of, say, 40 nm from the particle surface (although so long a distance is unnecessary), neglecting the small atomic dimensions we have $r = 0.06 \text{ µm} \ll r_{cr} = 4.53 \text{ µm}$. Even at $Q_{LPR} = 10$ —which is often the case in experiments— $r = 0.06 \ll r_{cr} = 1.34$, i.e., there is a safety margin of 1.5 orders of magnitude. This signifies that the retardation in Eqns (10)–(12) may be neglected.

Equations (10)–(12) can be generalized to describe DNLs of various designs: for instance, when there are N > 1 two-level systems (atoms) rather than one, each of the systems is located at a distance r_m from the nanoparticle center, as in Fig. 1b. Using the subscript *m* to mark the variables and parameters relating to the corresponding atom and introducing the variables $\overline{D} = N^{-1} \sum_{m=1}^{N} D_m$ and $\Sigma = N^{-1} \sum_{m=1}^{N} \Omega_m^* \sigma_m$ averaged over the atomic ensemble, where Ω_m is defined by expression (13) with $r = r_m$, one can



Figure 2. LPR quality factor as a function of the radius of a gold nanoparticle in water: I—result of an approximate solution to the dispersion equation [3], and 2—exact solution of this equation.

obtain, in lieu of formulas (10)-(12), the following equations

$$\dot{\bar{D}} = 2i(a^{+}\Sigma - \Sigma^{+}a) - \tau^{-1}(\bar{D} - D_{0}), \qquad (15)$$

$$\dot{\Sigma} = \left[\mathbf{i}(\delta_2 + \Omega_{dd}\bar{D}) - \Gamma_2 \right] \Sigma + \mathbf{i}|\bar{\Omega}|^2 a\bar{D} \,, \tag{16}$$

$$\dot{a} = (\mathrm{i}\delta - \Gamma_{\mathrm{LPR}})a - \mathrm{i}N\Sigma \,. \tag{17}$$

The factor N in the last term on the right-hand side of Eqn (17) points to the contribution made by N atoms to the generation of a nanoparticle's dipole moment. A peculiar feature of Eqn (16) is the nonlinear resonance shift $\Omega_{dd}\overline{D}$, where the quantity Ω_{dd} describing 'local field' effects in the DNL, which are associated with atomic interactions via the EMF [31], is defined by expression (13) averaged over all atoms, with r being the interatomic distance. It may be shown that Ω_{dd} is a real quantity. The inclusion of Ω_{dd} fluctuations permits describing the effects of inhomogeneous broadening ('self-broadening') in many-atom DNLs.

4. Oscillation conditions for the dipole nanolaser

We revert to Eqns (10)–(12). Dipole matrix elements $\mu_{0,2}$ enter into expression (13) for the coupling constant, but polarizabilities, whose calculation is carried out at will for metallic nanoparticles, are more convenient to use than $\mu_{0,2}$. In Ref. [10], it was shown that $|\mu_0|^2 = \alpha_{0r}\hbar\Gamma_{LPR}$ and $|\mu_2|^2 = \alpha_{2r}\hbar\Gamma_2$, where α_{0r} and α_{2r} are the respective resonance polarizabilities of the nanoparticle and the two-level system. Whence, and from expression (13), it is possible to obtain $|\Omega_{int}|^2/(\Gamma_2\Gamma) = |\xi|^2 |\alpha_{0r}\alpha_{2r}|/r^6$.

The nontrivial stationary solution of Eqns (10)–(12) is well known, in particular $D = D_{\text{th}} \equiv (1 + \delta^2 / \Gamma_{\text{LPR}}^2) \Gamma_2 \Gamma_{\text{LPR}} / |\Omega_{\text{int}}|^2$. Apart from the nontrivial solution, there is a trivial stationary solution $a = \sigma = 0$, $D = D_0$. When the pump exceeds the threshold value, $D_0 > D_{\text{th}}$, the trivial solution becomes unstable, and the nontrivial solution is realized, giving rise to the coherent generation of the dipole moment. Since the population of the upper state in the two-level system does not exceed unity, $D_{\text{th}} < 1$ becomes the necessary condition for the generation, which is fulfilled for a sufficiently strong interaction between the particles. In the case of reaching exact resonance $\omega = \omega_2 = \omega_{\text{LPR}}$, the necessary condition for DNL oscillation is given as follows

$$D_{\rm th}^{-1} = \frac{\left|\Omega_{\rm int}\right|^2}{\Gamma_2 \Gamma_{\rm LPR}} > 1$$

or

$$\xi|^2 \, \frac{|\alpha_{0r}\alpha_{2r}|}{r^6} > 1 \,, \tag{18}$$

i.e., the two-level system should be sufficiently close to the nanoparticle, so that their center distance *r* satisfies the inequality $r < r_{\rm cr} = |\xi|^{1/3} |\alpha_{0r}\alpha_{2r}|^{1/6}$.

The DNL threshold corresponds to the strongest interaction between the particles, i.e., according to inequality (18) to the maximum of the quantity $|\xi|^2/r^6$ in θ_r for a fixed r. An analysis made with the use of formulas (8) for a spherical nanoparticle suggests that the value of $|\xi|^2/r^6$ is greatest when $\cos \theta_r$ equals 1 or 0, i.e., when the dipoles are directed parallel or perpendicular to the segment connecting them; this also follows from symmetry considerations. The maximum at $\cos \theta_r = 1$ corresponds to $0 < k_2r < \rho_{\text{bif}}$, and in this case the near-field interaction of the dipoles prevails over their farfield interaction. The maximum at $\cos \theta_r = 0$ corresponds to



Figure 3. For a given value of *A*, the DNL oscillation is possible when $\rho < \rho_{\rm cr}(A)$, which is indicated with a solid curve. The curve break corresponds to $\rho_{\rm bif}$, on exceeding it, the direction of the dipoles becomes perpendicular to their connecting straight line. The magnitude of $A_{\rm Au}$ and the critical value of $\rho_{\rm Au}$ correspond to a DNL made of a spherical gold nanoparticle and a quantum dot in silicon.

 $k_2r > \rho_{\text{bif}}$ —the prevalent far-field interaction of the dipoles. At the point $k_2r = \rho_{\text{bif}} = (5 + \sqrt{37})^{1/2}/\sqrt{2} \approx 2.35$, the dipole interaction energies at $\cos \theta_r = 1$ and at $\cos \theta_r = 0$ are equal and $|\xi(\rho_{\text{bif}})|^2/\rho_{\text{bif}}^6 \approx 0.154$. Therefore, as the value of k_2r changes, in going across $k_2r = \rho_{\text{bif}}$ bifurcation occurs: a stepwise change in the direction of dipole polarizations and the DNL radiation directivity. If advantage it taken of the variable $\rho = k_2r$, the DNL oscillation condition (18) is conveniently written in dimensionless quantities:

$$A \frac{\left|\xi(\rho)\right|^{2}}{\rho^{6}} > 1,$$

$$\xi(\rho) = \begin{cases} 2 \exp(i\rho)(i\rho - 1), & 0 < \rho \le \rho_{\text{bif}}, \\ \exp(i\rho)(1 - \rho^{2} + i\rho), & \rho_{\text{bif}} < \rho < \rho_{\text{cr}} = k_{2}r_{\text{cr}}, \end{cases}$$
(19)

where $A = |\alpha_0 r \alpha_{2r}| k_2^6$. Figure 3 depicts the dependence $\rho_{cr}(A)$. The lasing is possible when $0 < \rho < \rho_{cr}$. In this case, when $A > A_{bif}$, two DNL oscillation modes are possible. The first is realized when $0 < \rho < \rho_{bif}$ and the dipoles are parallel to the segment connecting them, and the second when $\rho_{bif} < \rho < \rho_{cr}$ and the dipoles are perpendicular to the segment connecting them. Accordingly, the far-field radiation patterns of the dipoles in these modes will be rotated relative to each other: this property may be employed for controling the DNL radiation. Several oscillation modes and the corresponding bifurcations may also exist for nonspherical nanoparticles. For $\rho_{bif} < \rho < \rho_{cr}$, the degeneracy shows up in directions of the dipoles in the plane perpendicular to the segment connecting the particle centers; in this case, the atom must be treated as a three-level system rather than a two-level one.

Let us estimate the value of A for a gold spherical particle 7 nm in radius. Let us assume that the two-level system (molecule) resides on a dielectric shell of a nanoparticle in water; the refractive index of the shell is little different from water's refractive index $n_2 = 1.33$; these conditions are close to the experimental ones accepted in Ref. [25]. We utilize the parameters of the two-level system as in Ref. [10] and the same parameters and formulas for calculating the dielectric function of gold and the polarizability of a gold nanoparticle as in Ref. [34]. In this case, $\lambda_{LPR} = 525$ nm (in a vacuum), and $A = 6.28 \times 10^{-4}$, which corresponds to $\rho_{cr} = 0.378$. The greatest center-to-center distance of the particles is $r_{cr} =$ $\rho_{cr} \lambda_{LPR}/(2\pi n_2) = 24$ nm. In Ref. [25], for $\lambda_{LPR} = 520$ nm the distance between a shell surrounding a gold spherical particle of radius 7 nm and the center of this particle was equal to 22 nm, i.e., our estimates bear out the possibility of lasing in Ref. [25].

By way of a second example, let us consider a DNL comprising a gold (or silver) spherical nanoparticle of radius 10 nm and a quantum dot in silicon. By using the dielectric function of silicon [35], we find that $\lambda_{\text{LPR}} = 876$ (804) nm, A = 7.8 (6.7), and the dipoles may be spaced at a center-to-center distance $r < r_{\text{cr}} = 100$ (83) nm. The corresponding points for the gold nanoparticle are indicated in Fig. 3; for this particle, $A = A_{\text{Au}} > A_{\text{bif}} \approx 7$, and the dipoles may therefore be directed along as well as across their connecting straight line. These estimates suggest that the necessary condition for DNL oscillation can be fulfilled in real systems.

5. Inclusion of incoherent dipole nanolaser oscillation

In this section we consider, following report [14], DNL equations in the next approximation of the moments method, including the averages of quadratic quantities, which will permit describing the incoherent dipole moment oscillation—an analog of incoherent oscillation, including the subthreshold one, in an ordinary laser [29]. Under incoherent oscillation conditions, the average values of the dipole moments $\langle \sigma \rangle = \langle a \rangle = 0$, but the corresponding energies, or the squares of the moduli of the dipole moments, are $\langle \sigma^+ \sigma \rangle \neq 0$ and $\langle a^+ a \rangle \neq 0$.

In equations (10)–(12) we go over from operators a and σ to their binary combinations: the operator of the number of plasmons, $n_0 = a^+a$, and $G = i\Gamma_{tot}^{-1}(\Omega_{int}\sigma^+a - \Omega_{int}^*a^+\sigma)$, where $\Gamma_{tot} = \Gamma_{LPR} + \Gamma_2$. In lieu of D it is more convenient to use the operator n_2 of the upper level population of the two-level system: $D = 2(n_2 - 1/2)$. We put $\delta_0, \delta \ll \Gamma_{LPR}, \Gamma_2$ and consider the resonance case (which is more general than in Ref. [14]) and assume that there are N atoms equally spaced from a metal nanoparticle, for instance, on a dielectric shell (Fig. 1b). By differentiating the products of the operators and employing Eqns (10)–(12), we find that

$$\dot{G} = -2\Gamma_{\rm tot}G + 2g_{\rm pl}N\left[n_0\left(n_2 - \frac{1}{2}\right) + \frac{n_2}{2N}\right],$$
 (20)

$$\dot{n}_0 = -2\Gamma_{\rm LPR}n_0 + \Gamma_{\rm tot}G\,,\tag{21}$$

$$\dot{n}_2 = -\Gamma_{\rm tot}G - \frac{n_2}{\tau} + j\,,\tag{22}$$

where *j* is the rate of pumping of an individual atom, $g_{\rm pl} = 2|\Omega_{\rm int}|^2/\Gamma_{\rm tot} \equiv 4\Gamma_2\Gamma_{\rm LPR}/(D_{\rm th}\Gamma_{\rm tot})$, n_0 is the number of plasmons per atom, and the total number of generated plasmons is Nn_0 . In equations (20)–(22), the variables are the averages of the operators, and correlations are neglected, i.e., the averages are uncoupled: $\langle n_0 n_2 \rangle \approx \langle n_0 \rangle \langle n_2 \rangle$. In the derivation of equations (20)–(22), advantage was taken of the quantum-mechanical identity $\sigma^+\sigma = n_2$ [36], with the consequence that the term $n_2/(2N)$ responsible for the incoherent generation of a nanoparticle's dipole moment emerged in the square brackets on the right-hand side of Eqn (20). This term, which is proportional to n_2 , makes a contribution to plasmon generation irrespective of the existence of population inversion: the greater N, the smaller is this term.

Figure 4 depicts the dependence of the number of plasmons generated by a DNL with N = 100 atoms on the pumping rate expressed in Γ_{LPR} units at $D_{\text{th}} = 0.9$ and the same Γ_2 as in Ref. [10], i.e., $\Gamma_2 = \Gamma_{2\text{R}} + \Gamma_{2\text{NR}}$, where the



Figure 4. Number of plasmons generated by a DNL as a function of the pumping rate of each of N = 100 atoms, with the proviso that the population inversion be equal to 0.9 and the LPR linewidth be equal to 300 (curve 1), 30 (curve 2), and 3 (curve 3) atomic radiation linewidths. The threshold character of oscillation is evident in curve 3.

radiative width of the line of the two-level system (a quantum dot) is $\Gamma_{2R} \equiv 1/\tau = 1$ ns, the nonradiative linewidth $\hbar\Gamma_{2NR} = \bar{e}\gamma_{ac}T$, \bar{e} is the electron charge, T is the temperature [K], $\gamma_{ac} = 0.5 \times 10^{-6}$ eV K⁻¹; T = 300 K and $\Gamma_{LPR} = 300\Gamma_{2R}$ (curve 1), $\Gamma_{LPR} = 10\Gamma_{2R}$ (curve 2), and $\Gamma_{LPR} = 3\Gamma_{2R}$ (curve 3).

If the laser oscillation threshold is considered as a sharp acceleration of the growth in the number of plasmons upon increasing the pump current (the threshold in the number of plasmons), this threshold turns out to be evident only for curve 3, when the LPR Q factor is quite high. Curves 1 and 2 correspond to 'nonthreshold' lasing. No less important than the existence or the absence of the threshold in the number of plasmons are the conditions for oscillation line narrowing to a value smaller than Γ_{LPR} , which have not been investigated so far but may be found using the approach suggested in Ref. [30]. In view of the results of work [30], it may be assumed that inequality (18) remains the necessary condition for line narrowing even for 'nonthreshold' DNLs, which are described by curves like 1 and 2 in Fig. 4. It is significant that equations (20)-(22) predict a much higher pumping rate near the threshold: $j \sim \Gamma_{LPR}$ instead of $j \sim 1/\tau \ll \Gamma_{LPR}$ in the case of equations (10)-(12) disregarding the energy loss for incoherent subthreshold generation.

When $N \sim 1$, $\Gamma_{\text{LPR}} \ge \Gamma_2$ (a low-Q LPR), $\Gamma_{\text{tot}} \ge 1/\tau$, the number of plasmons $n_0 \ll 1$, and the pumping rate is not too high, $j \ll \Gamma_{\text{LPR}}$, it is possible to neglect the term $\sim n_0$ in Eqn (20) and adiabatically eliminate *G* from Eqns (20)–(22) by putting $\dot{G} = 0$. After this, only a single equation remains: $\dot{n}_2 = -(\tau^{-1} + g_{\text{pl}}/2)n_2 + j$. One can see from this equation that the electron lifetime in the upper state of the two-level system significantly shortens due to the presence of the metallic nanoparticle: under ordinary conditions $g_{\text{pl}} \sim \Gamma_{\text{LPR}} \ge \tau^{-1}$. This fact can be utilized for making broadband optical modulators with metallic nanoparticles [16].

In circumstances where $g_{pl} \sim \Gamma_{LPR} \gg \tau^{-1}$, upon adiabatic elimination of *G* it is possible to neglect the term n_2/τ in the equation for n_2 . This signifies that the atom radiates primarily to the nanoparticle's 'dipole' mode and that the radiation to its other modes with frequencies different from the dipole mode frequency is negligible. Therefore, the number of modes to which the atom may spontaneously radiate is limited, and in this sense the nanoparticle is similar to a photonic crystal. In this case, the stationary number of plasmons $n_0 = j/(2\Gamma_{\rm LPR})$ increases linearly with pump current, and there is no oscillation threshold in the number of plasmons, but the DNL linewidth may decrease to values smaller than Γ_{LPR} , as with a nonthreshold laser [30]. It is noteworthy that, although there is absorption in the metallic nanoparticle, the number of photons delivered by the twolevel system and reradiated by the particle is not so small in comparison with the number of photons emitted by the twolevel system in the absence of the nanoparticle, the pump rate being the same in both cases. Indeed, metallic nanoparticle cross sections for absorption and scattering under the LPR conditions may be in the ratio of 1:2 [4], i.e., only every third photon delivered by the atom will participate in nanoparticle heating, while the remaining photons will go to its dipole radiation. In this case, the reradiation rate is approximately $\tau \Gamma_{\text{LPR}}$ times (i.e., an order of magnitude or more) higher than the emission rate of the atom in the absence of the nanoparticle. Therefore, from the standpoint of converting the current of a high-power pump, the ensemble of atoms near the nanoparticle constitutes a highly efficient nano-dimensional light-emitting device.

6. Conclusions

The dipole nanolaser is a quantum electronic system remarkable for its basic properties and its possible practical applications. The uniqueness of the DNL lies, in particular, in the fact that it resides 'at the boundary' of the domain where quantum effects are significant, like spontaneous emission to an oscillation mode, as are classical nonlinear effects like the excitation of self-oscillations, etc. In the foregoing, it was impossible to consider all DNL-related quantum effects involving, for instance, superluminescence in DNL ensembles, oscillation line narrowing in overcoming the threshold, etc. Many classical nonlinear effects were not considered, either, like the DNL bistability in an external resonance field [14, 15]. Also of interest is to analyze a fewatom DNL-like a logic cell (a qubit) for a quantum computer, for instance, of the 'controlled NOT' type [37]. In the subthreshold generation, DNLs may be employed as efficient nanodimensional light emitters. To improve the efficiency of solar batteries and photodetectors, use can be made of a device 'inverse' to the DNL: when a nanoparticle — an optical antenna — receives a photon and transmits it via its near field to, for instance, a photocurrent-producing quantum dot. Extensive literature [7] is concerned with research aimed at improving the efficiency of solar elements with the help of metallic nanoparticles; the promise of quantum dot-based photoelectric converters, even without nanoantennas, is also recognized [38], but DNL photodetectors have not been considered so far. It is hard to foresee all promising DNL applications.

Different schemes of practical DNL realization are possible: notably, in the form of a metallic nanoparticle in a dielectric shell, on the surface of which the active atoms or molecules (for instance, dye molecules adsorbed from a solution with the nanoparticles) are located (Fig. 1b). This DNL scheme has been experimentally realized [25]. Calculations of this type of DNL can also be performed in the framework of the 'core–shell' nanoparticle model. The shells are treated as continuous media with corresponding (complex) refractive indices. Calculations for spherical nanoparticles can be performed using the well-known modes of a spherical cavity [3] without invoking the quasiclassical approximation. Also possible is the employment of a 'nanoparticle-two-level system' model, similar to the model presented above, with several radiating atoms (molecules). Another DNL scheme incorporates a nanoparticle antenna placed on the surface, say, of a semiconductor; located at a small (up to several dozen nanometers) depth below the surface is an active layer—a p-n junction, a quantum well, or a layer of quantum dots. There are examples of the practical implementation of such systems, and the enhancement of their electro- and photoluminescence due to metallic nanoparticles is well known [23, 24]. A theoretical description of this type of DNL must take into consideration the plane interfaces of the media and the electrodynamics of dipoles in stratified media [39]. Also conceivable are combinations of various DNL schemes, for instance, of metallic nanoparticles with active shells residing on semiconductor surfaces.

The DNL theory, relying on rather simple equations, is developing systematically, providing a way for describing complex DNL schemes, predicting new effects, and helping in the planning of experiments. This enables the researcher to make analytical estimates using the results of theoretical quantum electronics, plasmonics, and the theory of radiation-matter interactions. Next, by applying numerical techniques, it will be possible to carry out comprehensive simulations of experiments, as soon as they are planned in Russia.

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Quantum plasmonics of metamaterials: loss compensation using spasers

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1. Introduction

Recent years have seen the development of a new field of optics-quantum plasmonics-which combines the advantages of plasmonics and quantum electronics [1-25]. Although plasmonics deals with wave phenomena, it operates on a scale much shorter than the light wavelength in a vacuum, which endows plasmonics with many features of near-field optics and creates a demand for plasmonics from modern nanotechnologies. In the first place, mention should be made of SERS (surface enhanced Raman scattering), the SPASER (surface plasmon amplification by stimulated emission of radiation), nanodimensional light sources [26– 30], and numerous metamaterial-based devices [17, 31, 32]: energy concentrators and transmission lines on the order of several dozen nanometers in size, a superlens with a resolution exceeding the diffraction limit, cloakings, hyperlenses [33-40], etc. The small dimensions of these objects introduce quantum effects into their dynamics.

Since the principle of metamaterials operation is underlain by the plasmon resonance of metallic nanoparticles (NPs), artificial metamaterials exhibit rather high energy loss. The existence of losses in metamaterial-based devices

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Uspekhi Fizicheskikh Nauk **182** (10) 1122–1130 (2012) DOI: 10.3367/UFNr.0182.201210j.1122 Translated by E N Ragozin; edited by A Radzig gives rise to energy transfer inside of them, which is effected by near fields. The necessary and sufficient condition for the energy transfer by evanescent waves is the emergence of a phase difference among 'interfering' evanescent harmonics [41]. The emerging dephasing of harmonics, which form an ideal image, shows up in their destructive interference and breaking of the ideal image [42]. To compensate for the loss, the authors of Refs [43–51] proposed the employment of active (amplifying) media in artificial metamaterials. However, it follows from the foregoing that the ideal image is broken not only by energy dissipation, but also amplification in the medium. It is required that as precise as possible a loss compensation be achieved [43, 52, 53].

The utilization of active media in metamaterials leads inevitably to the formation of nanolasers inside of them. Among nanolasers, mention should be made of the dipole nanolaser [8, 10], the spaser [11, 54], and the magnetic-mode nanolaser [48, 49]. From the standpoint of loss compensation in metamaterials, spasers, whose experimental realization was reported in Ref. [55], have the greatest promise as a base element. Schematically, the spaser constitutes a quantumplasmon device which consists of inversely excited two-level quantum dots (ODs) (a two-level tunneling system, TLS) surrounding plasmon NPs (the more realistic treatment of a four-level system does not introduce qualitatively new properties (see Refs [50, 56, 57])). The principle of spaser operation is similar to that of lasers: light amplification ensured by population inversion in combination with feedback, which is produced by the stimulated emission of a quantum system. To fulfill the conditions for stimulated emission by an inverted quantum system in the field of the wave previously radiated by this system, the quantum system is placed in a cavity, which localizes the generated mode. In a spaser, the role of photons is played by surface plasmons (SPs) of an NP. The localization of plasmons on the NP [11, 49, 54] furnishes the conditions for feedback realization. To state it in different terms, the generation and amplification of the NP's near-fields occur in spasers. The amplification of SPs proceeds due to radiationless energy transfer from QDs. The process relies on the dipole-dipole interaction (or any other near-field interaction [58]) between a QD and a plasmon NP. This mechanism can be treated as the principal one, because the probability of radiationless plasmon excitation is $(kr_{\rm NP-TLS})^{-3}$ times higher than the probability of radiative photon emission [15] (r_{NP-TLS}) is the center-to-center distance of the NP and the QD, $k = 2\pi/\lambda$, where λ is the wavelength in a vacuum). Therefore, the efficient energy transfer from the QD to the NP is achieved due to the short distance between them, despite the fact that the plasmon resonance Q factor is rather low.¹ Due to the high efficiency of this process, an external optical wave which propagates through the metamaterial interacts with entire spasers rather than separately with the amplifying medium and separately with plasmon particles.

Like a laser, a spaser constitutes a self-oscillating system. Its dipole moment executes free-running oscillations whose frequency and amplitude are determined by the balance between pumping and dissipation. An external field can only synchronize the spaser operation, i.e., make the dipole moment oscillate at the frequency of the external field. The weak dependence of the amplitude of these oscillations on the

¹ Below, we neglect the emission of photons, and therefore the Purcell effect [59, 60] does not play an appreciable role and may be disregarded.