PHYSICAL REVIEW A **71**, 063812 (2005)

Dipole nanolaser

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(Received 6 December 2004; published 17 June 2005)

A "dipole" laser is proposed consisting of a nanoparticle and a two-level system with population inversion. If the threshold conditions are fulfilled, the dipole interaction between the two-level system and the nanoparticle leads to coherent oscillations in the polarization of the particles, even in the absence of an external electromagnetic field. The emitted radiation has a dipolar distribution. It does not need an optical cavity, and has a very small volume, $\sim 0.1\ \mu\mathrm{m}^3$, which can be important for applications in microelectronics. Estimates of the threshold conditions are carried out for a dipole laser composed of a quantum dot and a silver nanoparticle.

DOI: 10.1103/PhysRevA.71.063812 PACS number(s): 42.55.Ah, 42.70.Nq, 42.70.Hj

I. INTRODUCTION

It is well known that an admixture of nanoparticles may significantly change the optical properties of a medium. For example, the enhancement of the spontaneous emission from an atom next to a metallic nanoparticle has been studied both theoretically and experimentally [1]. Similarly, enhanced fluorescence intensities have been observed and studied in fluorophores near metallic nanostructures and surfaces, including enhanced rates of multi-photon excitation, and clearly modified rates of energy transfer [2]. Spectral hole-burning and fluorescence enhancement were observed in MgS:Eu nanoparticles [3]. Subwavelength patterning of the optical near field has been observed in a regular array of resonant gold nanoparticles [4], whereas modification of the optical properties of photonic crystals by metallic nanoparticles have been studied and observed in [5]. The change in the optical properties of a medium caused by metallic nanoparticles of various shapes because of dipole-dipole interactions between the particles was studied in [6], and features in the light absorption of silica nanoparticles were reported in [7].

There is also high interest in the construction and study of miniature nanolasers based on photonic crystals [8] or nanowires [9] with very small optical cavities. Nanolasers are considered to be one of the critical building blocks for nanoscale optoelectronics [10].

Despite their small volumes, the nanolasers proposed thus far all work in the same way as conventional lasers. In particular, to provide feedback between the lasing field and the active medium they must have an optical cavity whose size, at least along one dimension, is comparable to or greater than the laser wavelength in the medium. Here we show that nanoparticles may bring a different way of lasing. We propose a nanolaser incorporating a metallic nanoparticle and a two-level system (TLS): this laser does not need an optical cavity [11]. The proposed nanolaser utilizes coupling through the near (local) field between a resonant transition of the active particle and the plasmon resonance of the metallic nanoparticle. Such coupled interactions are known to strongly modify optical emission. They are, for instance, responsible for surface-enhanced Raman scattering, where the plasmon resonance due to a regular array of metallic nanoparticles can lead to strong resonant emission, as reported, for example, in [12].

A physical understanding of the proposed lasing mechanism can be obtained by noting that, under certain conditions, metallic nanoparticles can act as high-quality "antennas" for active atoms, leading to an increase in the amplification coefficient of an active medium [13]. How can this lead to lasing? Consider first the usual lasing mechanism. This involves stimulated emission into a mode (modes) of a cavity from a medium in which there is a population inversion [14]. When the threshold conditions are satisfied so that the stimulated emission exceeds the stimulated absorption and the losses, then energy is transferred from the incoherent pump into coherent laser radiation. Because of the Bosonic nature of the electromagnetic field, all emitted photons have almost the same wave vector, close to the wave vector of the cavity mode, leading to a narrow lasing spectrum with high power.

Besides the electromagnetic field, Bose operators can describe any harmonic oscillations, including, for example, linear oscillations of the charge and polarization of a medium. By exciting the medium in such a way that the total energy flux into the polarization exceeds the losses, one can obtain, in principle, polarization oscillations with a narrow spectrum which lead, in turn, to lasing. We propose here a *dipole nanolaser* (DNL), where coherent polarization oscillations of a nanoparticle can be excited through its dipole interaction with an inverted TLS based, for example, on a quantum dot or an atom.

The minimum volume of conventional miniature lasers, such as VCSELs [15], micro-sphere lasers [16], or the nanolasers mentioned above, is restricted by the volume of a cavity mode, which is, typically, greater than the cube of the lasing wavelength λ. Because the DNL uses near-field interactions, the DNL does not need an optical cavity and may have a volume $\ll \lambda^3$, which could be of benefit for a wide range of applications, including microelectronics.

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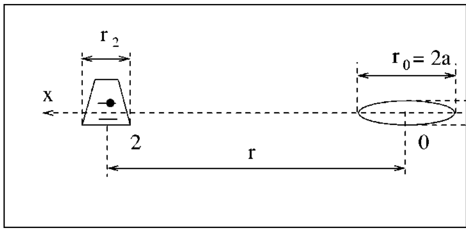

FIG. 1. Scheme of dipole nanolaser. Parameters of the nanoparticle have index 0, parameters of the two-level system have index 2.

Surface-plasmon amplification by stimulated emission of radiation (spaser) has been proposed in [17]. The spaser approach has some features in common with the DNL. In particular, the spaser does not need an optical cavity. The spaser, however, operates in a regime that generates near-field modes, while the DNL generates polarization. The DNL approach could be referred to as "polarization amplification by stimulated emission of radiation" (PASER). However, the name DNL emphasizes that we are interested, above all, in the emission of strong and coherent radiation by a nano-object, and it avoids any confusion between the polarization of the medium and the polarization due to the electromagnetic field. A comparison between the DNL and the spaser will be carried out in Sec. III.

Let us now consider a rather simple model of the DNL, to explain its operation, and to estimate the working conditions and parameters for a DNL.

II. EQUATIONS OF MOTION

Suppose that a two-level system (TLS) of size r_2 is placed at a distance r from a nanoparticle of size r_0 in a solid dielectric or semiconductor as shown in Fig. 1. An incoherent pump provides a population inversion in the TLS. The pump may be, for example, a broadband optical pump to a higher level of the TLS, as in a three-level laser pump scheme, or may be provided by carrier injection from the bands of a semiconductor material surrounding an embedded quantum dot.

We use a quantum-mechanical derivation of the equations of motion for the system shown in Fig. 1, but neglecting quantum correlations and fluctuations in the analysis. The Hamiltonian of the nanoparticle and TLS is given by

$$H = H_0 + H_2 + \hat{V} + \hat{\Gamma},$$

where H_0 and H_2 describe, respectively, the nanoparticle and TLS, ignoring their interaction. The operator $\hat{V}$ gives the interaction between the TLS and the nanoparticle, while $\hat{\Gamma}$ includes the terms describing dissipation and pump effects.

Free electrons in the nanoparticle can oscillate with a frequency ω_0 near to the plasmon resonance, leading to harmonic oscillations of the nanoparticle dipole moment $\mu_0[a(t)+a^+(t)]$. Here μ_0 is the matrix element of the dipole moment operator of the nanoparticle, and $a(t)$ is a Bose operator. Suppose that an electron in the TLS makes transitions between the high and the low TLS states, so that the dipole moment of the TLS oscillates with a frequency $\omega \approx \omega_2$, where ω_2 is the transition frequency of the TLS. Because of the interaction $\hat{V}$, the electrons in the nanoparticle will also oscillate with the same frequency ω. Supposing that $\omega \approx \omega_0 \approx \omega_2$, we can write equations of motion for the slowly varying amplitude operators $a_0(t) \equiv a(t)e^{i\omega t}$ and $\sigma(t) \equiv \varrho(t)e^{i\omega t}$, where $\varrho(t)$ is the operator describing transitions between the low and high states of the TLS. $D(t)=n_a(t)-n_b(t)$ is the population inversion operator, with n_a (n_b) operators for the population of the high (low) state of the TLS. Neglecting the fast oscillating terms $\sim e^{\pm 2i\omega t}$, we can write

$$H_0 = \hbar\omega_0 a^\dagger a, \quad H_2 = \hbar\omega_2 n_a,$$

$$\hat{V} = \hbar\Omega_{\text{int}}(a_0^\dagger\sigma + \sigma^\dagger a_0). \tag{1}$$

In order to simplify the analysis we assume that

$$r_0, r_2 \ll r \ll k^{-1}, \tag{2}$$

where $k=\omega_2/c$ is the wave number associated with photons emitted and/or absorbed by an isolated TLS. If the conditions in Eq. (2) are satisfied, then

$$\Omega_{\text{int}} = [\vec{\mu}_0 \,.\, \vec{\mu}_2 - 3(\vec{\mu}_0 \,.\, \vec{e}_r)(\vec{\mu}_2 \,.\, \vec{e}_r)]/(\hbar r^3), \tag{3}$$

which is the matrix element of the static dipole-dipole interaction operator [18], where the components of $\vec{\mu}_2$ are matrix elements of the dipole moment operator of the TLS resonant transition, and $\vec{e}_r$ is a unit vector directed from the nanoparticle to the TLS. The sum $n_a+n_b=n_0$, where $0<n_0\leq 1$ is a constant. If the TLS is a quantum dot, then n_0 is determined from the equilibrium conditions associated with the carrier distribution in the TLS and the semiconductor medium around the TLS.

By using the well-known commutation relations for the operators a_0, σ, and $n_{a,b}$ [19] we can derive the equations of motion

$$\dot{D} = 2i\Omega_{\text{int}}(a_0^\dagger\sigma - \sigma^\dagger a_0) - \frac{D - D_0}{\tau} \tag{4}$$

$$\dot{\sigma} = (i\delta - \Gamma)\sigma + i\Omega_{\text{int}}a_0 D \tag{5}$$

$$\dot{a}_0 = (i\Delta - \Gamma_0)a_0 - i\Omega_{\text{int}}\sigma, \tag{6}$$

where $\delta=\omega-\omega_2$, $\Delta=\omega-\omega_0$. We have included in Eqs. (4)–(6) terms involving $1/\tau$, Γ and Γ_0 to describe the relaxation and pump processes. D_0 is the stationary value of D when $\sigma=a_0=0$, with

$$\frac{1}{\tau} = \frac{1}{\tau_d} + \frac{1}{\tau_p}, \quad D_0 = n_0\frac{\tau_d - \tau_p}{\tau_d + \tau_p}, \tag{7}$$

where $1/\tau_d$ and $1/\tau_p$ are, respectively, the damping and the pumping rates. We suppose $\tau_p<\tau_d$ and therefore $D_0>0$, so that the pump provides a population inversion in the TLS. We neglect quantum fluctuations and correlations, so that D, σ, and a_0 can be treated as complex variables with σ^+ and a_0^+ then replaced by σ^* and a_0^*, respectively.

III. THRESHOLD CONDITIONS AND THE LASING RATE

Equations (4)–(6) are identical to the Maxwell-Bloch equations for a TLS in the electromagnetic field of a cavity mode with Rabi frequency Ω_{int}. It is well-known that these equations have not only the trivial stationary solution $a_0=0$, $\sigma=0$, $D=D_0$, but also the nontrivial stationary solution

$$D = D_{th} \equiv \frac{\Gamma\Gamma_0}{\Omega_{\text{int}}^2}\left(1 + \frac{\Delta^2}{\Gamma_0^2}\right), \tag{8}$$

$$\delta/\Gamma = -\Delta/\Gamma_0 \tag{9}$$

$$a_0 = \frac{e^{i\varphi}}{2}\left[\frac{D_0 - D_{th}}{\Gamma_0\tau}\right]^{1/2}, \quad \sigma = a_0\frac{\sqrt{\Gamma_0^2+\Delta^2}}{\Omega_{\text{int}}}e^{i\Delta\varphi}, \tag{10}$$

$$e^{i\Delta\varphi} = \frac{\Delta + i\Gamma_0}{\sqrt{\Gamma_0^2+\Delta^2}},$$

where φ is an arbitrary constant phase. Because $D \leqslant D_0 \leqslant 1$, the nontrivial stationary solution has physical sense if $D < 1$, which requires, according to Eq. (8), that

$$\frac{\Omega_{\text{int}}^2}{\Gamma\Gamma_0} > \left(1 + \frac{\Delta^2}{\Gamma_0^2}\right). \tag{11}$$

When condition (11) is true and $D_0 > D_{th}$, the stationary solution (8)–(10) can exist, giving a nonzero dipole moment for the nanoparticle and for the TLS. Note that these nonzero dipole moments (or polarization) appear even in the absence of an external electromagnetic field. By direct analogy with the results of laser theory [14], we can therefore predict that the spectral width of the polarization oscillations will decrease and become smaller than Γ and Γ_0 with increasing $|a_0|$.

Because of the coherent oscillations of the polarization, the nanoparticle and TLS will emit coherent dipole radiation of frequency ω. When the conditions of Eq. (2) are true, the spatial pattern of the radiation from the particles is the same as that due to a single dipole. The spectrum of that dipole radiation will tend to narrow with increasing $|a_0|$. We note that this radiation enters in Eqs. (8)–(10) only through the contribution of the radiation losses to the relaxation rates $1/\tau$, Γ and Γ_0 (see Sec. IV). In Eqs. (8)–(10) the polarization of the nanoparticle ($\sim a_0$) replaces the electromagnetic field in the usual laser theory. We refer to the phenomena described by the nontrivial stationary solution of Eqs. (8)–(10) as *dipole lasing*.

Equations similar to (8)–(10) can be derived, in principle, through an extension of the formalism used for the spaser in [17]. This can be done through adiabatic elimination, under proper conditions, of all the surface plasmon modes [see Eq. 4 of [17]], to obtain the interaction energy between the TLS and the metallic nano-object and so to derive the equations of motion. In this way the theoretical approach taken to describe the spaser [17] can also be used for the DNL, and the threshold condition can be found for the spontaneous appearance of polarization [similar to Eq. (11)]. This threshold condition may be different from the threshold condition for a particular surface-plasmon (SP) mode, given by Eq. 5 of [17]. Indeed, the near field in the DNL includes all SPs with $\omega \approx \omega_0 \approx \omega_2$ contributing into the interaction of the TLS with the nanoparticle. Free-space emission from the DNL, as well as the strong SP modes, appear as a consequence of a spontaneous buildup of the nanoparticle polarization. Whether a spaser or DNL is realized depends on which lifetime is longer: the SP lifetime τ_{near} or the coherent polarization lifetime τ_{pl} of the nanoparticle at the plasmon resonance. In the case of the dipole interaction considered in this paper, $\tau_{\text{near}} \sim r/c \leqslant \omega^{-1}$. Indeed, the microscopic mechanism for the dipole interaction is the emission of a photon by the TLS, the absorption of that photon by the nanoparticle and then photon emission into free space [29]. Therefore $\tau_{pl}=Q/\omega \gg \tau_{\text{near}}$, where $Q \gg 1$ is the quality factor for nanoparticle polarization oscillations at the plasmon resonance. When the TLS is close to nano-objects of complex structure, as in [17], the contribution of all multipole terms into the near-field interaction becomes important, so that one may reach the limit $\tau_{pl} \ll \tau_{\text{near}}$, which is the case for the spaser.

The stationary solution of Eqs. (8)–(10) is most easily realized when the inversion conditions are minimized, which requires, from Eq. (8), to maximize Ω_{int}. As one can see from Eq. (3), Ω_{int} reach its maximum when $\vec{\mu}_0$ is parallel to $\vec{\mu}_2$. The dipole moment μ_0 of the ellipsoidal nanoparticle shown in Fig. 1 has its maximum value along the x axis, and therefore the magnitude of Ω_{int} is maximized when $\vec{\mu}_0$ and $\vec{\mu}_2$ are parallel and directed along this axis.

Let us find the lasing rate γ_R describing the rate of dipole radiation from the TLS and from the nanoparticle. For the conditions given by Eq. (2), the total dipole moment of the TLS and the nanoparticle is the sum of the dipole moments of each particle, so that the dipole radiation rate [18] is

$$\gamma_R = \frac{4k^3}{3\hbar}|d_0 + d_2|^2, \tag{12}$$

where $d_0=2\mu_0 a_0$, $d_2=2\mu_2\sigma$, and a_0 and σ are given by Eqs. (8)–(10). It is convenient to express γ_R in terms of the resonant polarizabilities $\alpha_0(\omega)$ and $\alpha_2(\omega)$ of the nanoparticle and the TLS

$$\alpha_0 = \alpha_{0r}\frac{i - \bar{\Delta}}{1+\bar{\Delta}^2} \quad \alpha_2 = \alpha_{2r}\frac{\bar{\delta} - i}{1+\bar{\delta}^2}, \tag{13}$$

where $\alpha_{0r}=\mu_0^2/(\hbar\Gamma_0)$, $\alpha_{2r}=\mu_2^2/(\hbar\Gamma)$, $\bar{\Delta}=\Delta/\Gamma$, and $\bar{\delta}=\delta/\Gamma$. Equations (13) follow from Eqs. (5) and (6), if one replaces the terms $\sim\Omega_{\text{int}}$ by $-\mu_2 E/\hbar$ in Eq. (5) and by $-\mu_0 E/\hbar$ in Eq. (6), where E is the amplitude of an external electromagnetic field. By inserting the stationary solutions (10) into Eqs. (11), (12), and (8) and by using Eq. (13), we derive

$$\gamma_R = \frac{4k^3\alpha_{0r}}{3\tau}(D_0 - D_{th})\left|1 + \frac{r^3}{2|\alpha_0|}e^{i\Delta\varphi}\right|^2, \tag{14}$$

$$D_{th} = \frac{r^6}{4|\alpha_0||\alpha_2|} \leqslant 1, \quad D_{th} < D_0 \leqslant 1. \tag{15}$$

Note that the TLS polarizability α_2 enters only in D_{th}, [see Eq. (15)], while the nanoparticle polarizability α_0 also appears in Eq. (14) for γ_R. This implies that the radiation is caused, at most, by polarization oscillations of the nanoparticle.

If we add terms $\mu_2 E/\hbar$ and $\mu_0 E/\hbar$ into Eqs. (5) and (6) and find the stationary solution, we can obtain the polarizability α_{tot} of the TLS and the nanoparticle in a monochromatic field E. This case will be analyzed in detail elsewhere. Here we only note that $\alpha_{\text{tot}} \to \infty$ when $D_0 = D_{th}$, implying the spontaneous appearance of nonzero dipole moment when the external electromagnetic field $E \to 0$. One can see that this is in direct analogy with the linear laser amplifier, where the amplification coefficient goes to infinity at the lasing threshold (see, for example [20]). On the other hand, it is also well known that the linear DC polarizability goes to infinity for a ferroelectric phase transition [21]. Besides, the theory of the local field in a dielectric medium also predicts $\alpha_{\text{tot}} \to \infty$ under certain conditions, giving the so-called Clausius-Mossotti catastrophe [22]. The similarities among dipole lasing, an ordinary laser, ferroelectric phase transitions, and local field phenomena will be discussed in more detail elsewhere.

With the help of Eqs. (14) and (15) we can estimate the magnitude of the nanoparticle and TLS parameters required in order to achieve dipole lasing. Toward this end, we determine in the Sec. IV the polarizability of the nanoparticle in terms of the generally used expressions for the dielectric functions ϵ and ϵ_m of the materials of the nanoparticle and the surrounding "matrix" medium.

IV. ESTIMATION OF POLARIZABILITY OF THE NANOPARTICLE

In Eq. (6) $\Gamma_0 = \Gamma_T + \Gamma_R$, where Γ_T is the relaxation rate related to thermal and other nonradiative losses, and Γ_R is the radiative loss rate. We define $\gamma \equiv \Gamma_R/\Gamma_T$ to be the ratio of the two relaxation rates. We then rewrite Eq. (13) for α_0 identically as

$$\alpha_0 = \alpha_T\left(1 - \frac{\gamma}{1 + \gamma - i\overline{\Delta'}}\right), \qquad (16)$$

where $\alpha_T = i\alpha_{T0}/(1 - i\overline{\Delta'})$, $\overline{\Delta'} = \Delta/\Gamma_T$, and $\alpha_{T0} = \mu_0^2/(\hbar\Gamma_T)$. The radiative loss rate for the polarization amplitude is $\Gamma_R = (2/3)\mu_0^2 k^3/\hbar$, and therefore $\gamma = (2/3)\alpha_{T0}k^3$. In the limit $\gamma \to 0$ we have $\alpha_0 \to \alpha_T$ and α_T approaches the static polarizability of the nanoparticle. Following Ref. [27], the static polarizability of an ellipsoidal nanoparticle is

$$\alpha_{st} = \frac{V}{4\pi}\frac{\tilde{\epsilon} - 1}{1 + n(\tilde{\epsilon} - 1)}, \qquad (17)$$

where V is the volume of the nanoparticle, $0 < n < 1$ is the depolarizability factor, which depends on the ratio a/b of the nanoparticle semiaxes and $\tilde{\epsilon} = \epsilon/\epsilon_m$, where ϵ and ϵ_m are, respectively, the dielectric functions of the nanoparticle material and of the surrounding medium. By equating $\alpha_T = \alpha_{st}$ near the plasmon resonance we find

$$\alpha_{T0} = \frac{V}{4\pi}\left[\tilde{\epsilon}'' + \frac{(\tilde{\epsilon}' - 1)^2}{\tilde{\epsilon}''}\right], \quad \overline{\Delta'} = -\frac{\tilde{\epsilon}' - 1}{\tilde{\epsilon}''}[n(\tilde{\epsilon}' - 1) + 1], \qquad (18)$$

where $\tilde{\epsilon}' = \epsilon'/\epsilon_m$, $\tilde{\epsilon}'' = \epsilon''/\epsilon_m$, $\epsilon = \epsilon' + i\epsilon''$ are taken at the plasmon resonance frequency, and ϵ_m is real. We have, finally, $\alpha_{0r} = \alpha_{T0}/(1+\gamma)$ and $\overline{\Delta} = \overline{\Delta'}/(1+\gamma)$, so that all parameters in Eq. (13) for α_0 are expressed through well-known quantities. Because $\alpha_T = \alpha_{st}$ only at $\gamma = 0$, expression (18) gives good quantitative results only for $\gamma \ll 1$, which is usually the case. A rigorous calculation of α_0 can be carried out in the frame of Mie theory [23].

V. ESTIMATION OF NECESSARY CONDITION FOR DIPOLE LASING

Let us take the resonant wavelength $\lambda = 1.3$ μm and suppose that the nanoparticle and TLS are embedded in a semiconductor "matrix" medium with $\epsilon_m = 11.7$ [24]. We assume a silver ellipsoidal nanoparticle with $\epsilon'(\lambda) = -55$ and $\epsilon''(\lambda) = 5.74$ at $T_0 = 300$ K [25]. If the temperature T is of the order of tens of Kelvin or more, then $\epsilon''(\lambda) \sim T$ according, for example, with the Drude model [26], so that we can estimate $\epsilon''(T) = 5.74(T/300)$. We suppose that the ratio of the axes of the nanoparticle is $a/b \approx 2$, so that the depolarizability factor $n \approx 0.175$ [27]. By inserting these values into the second equation of Eqs. (18) we find $\overline{\Delta} = 0.027$, which means that $\lambda = 1.3$ μm is very close to the plasmon resonance of the nanoparticle. We take the nanoparticle axes to be of length $2a = 13$ nm and $2b = 2c = 6.5$ nm; the volume of the nanoparticle is $V = (4/3)\pi ab^2$. We consider a quantum dot of size $2r_2 = 10$ nm as the TLS. By using the expression for the radiative rate of the TLS, $\Gamma_{2R} = (4/3)(\mu_2^2 k^3/\hbar)$, where $k = 2\pi/\lambda$ we can represent the resonant polarizability of the TLS as

$$\alpha_{2r} = \frac{3}{4k^3}\frac{\Gamma_{2R}}{\Gamma}, \quad \Gamma = \Gamma_{2R} + \Gamma_{2T}(T), \qquad (19)$$

where $\Gamma_{2T}(T)$ is the width of the nonradiative broadening. The homogeneous linewidth of a single quantum dot was studied in [28], where it was found that at $T < 60$ K, $\Gamma_{2R} \approx 1$ ns, and $\Gamma_{2T}(T) \approx \gamma_{ac}T$, where $\gamma_{ac} \sim 0.5$ μeV K^{-1}.

Dipole lasing is possible if $D_{th} \leqslant 1$, which requires, according to Eq. (15), that the distance r between the TLS and the nanoparticle must be small

$$r \leqslant r_{cr} = (4|\alpha_0||\alpha_2|)^{1/6}. \qquad (20)$$

Because the particles have finite sizes, we also require that $r_{cr} > r_2 + a$ (see Fig. 1). From Eq. (2), the condition for the dipole approximation to be valid is that $r_{cr} - (r_2 + a) \gg \max(2r_2, 2a)$. Figure 2 shows the dimensionless parameter $\xi \equiv [r_{cr} - (r_2 + a)]/2a$ as a function of T with $r_{cr}(T)$ determined by Eq. (20). As one can see from the dotted curve in Fig. 2, the necessary condition for dipole lasing $\xi > 0$ can be satisfied even if we neglect the decrease of ϵ'' with temperature for $T < 300$ K. The condition $\xi \gg 1$ necessary for the validity of the dipole approximation is more or less satisfied

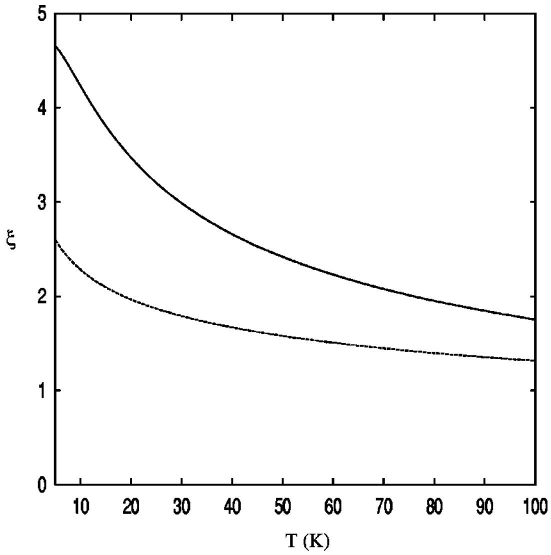

FIG. 2. Dimensionless distance ξ between the TLS and the nanoparticle as a function of temperature. The necessary condition for dipole lasing is $\xi > 0$. The dipole approximation used in our estimates is valid if $\xi \gg 1$. The solid line assumes $\epsilon'' \sim T$, the dashed line uses the $T=300$ K value of $\epsilon''=5.74$.

at low temperatures (few tens of Kelvin), assuming a linear decrease of ϵ'' with temperature. The condition $r \ll k^{-1}$ [see Eq. (2)] is satisfied, indeed $r \leqslant 4\times 2a = 52$ nm $\ll k^{-1} \approx 210$ nm. Thus, we conclude that the necessary condition for dipole lasing can, in principle, be satisfied. The conditions will be most easily satisfied with a narrow plasmon resonance. More detailed studies of dipole lasing are also required to take into account the interaction between particles beyond the dipole approximation.

VI. DISCUSSION

Let us summarize the physical mechanism and some particular features of dipole lasing. The TLS and the nanoparticle are placed close to each other, so that there is a strong interaction (in particular, a dipole interaction) between them through the near field. The condition of Eq. (11) (at $\Delta=0$) that $\Omega_{\mathrm{int}} > \sqrt{\Gamma\Gamma_0}$ means that the interaction rate must exceed the average polarization loss rate. The near field then provides the feedback between the nanoparticle and the TLS, which stimulates coherent oscillations of their dipole moments and leads to coherent dipole radiation into the surrounding space. The lifetime of a photon in the near field is very small, $\sim r/c$, where c is the speed of light. This is why the near field can be adiabatically eliminated from the treatment (as it is in [29]), which leads to the Hamiltonian of Eq. (1) with the interaction term $\hat{V}$ [see Eqs. (1) and (3)].

The spatial pattern of dipole nanolaser radiation is the same as that for a single dipole. Far above the lasing threshold, the radiation linewidth can be narrower that Γ and Γ_0. In the ideal case the dipole lasing linewidth is determined by quantum fluctuations of the nanolaser dipole moment $d_0 + d_2$, just as the linewidth of an ideal conventional laser is determined by quantum fluctuations in the lasing mode. In practice, the linewidth of the dipole nanolaser will increase due to thermal and other noise factors, which can be taken into account by adding appropriate Langevin forces into Eqs. (4)–(6).

Let us compare the threshold conditions for a dipole nanolaser with the threshold conditions for an ordinary laser. These conditions can be obtained from Eqs. (4)–(6), where a_0 is a dimensionless measure of the amplitude of the lasing mode and we replace Ω_{int} by $\Omega_R = \mu_2 E_R/\hbar$, and where $E_R = \sqrt{2\pi\hbar\omega/V_m}$ and V_m is the effective volume of the lasing mode. By setting $\Omega_{\mathrm{int}} = \Omega_R$ we find that the dipole nanolaser is comparable to an ordinary laser for which the volume of the lasing mode is

$$V_m = \frac{\pi r^6}{2\alpha_{0r}} Q_0, \tag{21}$$

where $Q_0 = \omega/\Gamma_0$ is the quality factor for polarization oscillations in the nanoparticle. From the Drude model we estimate $Q_0 \sim 50$ for our case. Thus when the distance between the particles, $r \sim 3\times 2a \sim 50$ nm, the dipole nanolaser corresponds to an ordinary laser with $V_m \approx 0.17\ \mu\mathrm{m}^3$. The small effective volume of a dipole nanolaser arises because the nanoparticle plays the role of the cavity, and the interaction between the TLS and the nanoparticle occurs through *near-field modes*, which are different from the electromagnetic wave-cavity modes of an ordinary laser. The small value of V_m allows one to satisfy the threshold conditions, even for rather small $Q_0 \sim 50$, and therefore to propose a typical size of a dipole nanolaser of the order of a few tens of nanometers.

If the cavity escape losses dominate in an ordinary laser, then the lasing rate is equal to the pump rate of the TLS

$$\gamma_{\mathrm{las}} \equiv 2\Gamma_0 |a_0^\dagger e^{i\omega t} + a_0 e^{-i\omega t}|^2 = (1/\tau)(D_{th} - D_0).$$

With the help of Eq. (14) one can see that γ_R is different from γ_{las} by a factor $\sim k^3\alpha_{0r}$, if $|\alpha_0|/r^3 \gg 1$, or by a factor $\sim (kr)^3$, if $|\alpha_0|/r^3 \leqslant 1$. By taking into account that $\alpha_{0r} \sim \alpha_{0s}/(1+\gamma)$, we conclude that in either case $\gamma_R \leqslant \gamma_{\mathrm{las}}$.

The threshold condition of Eq. (15) can be interpreted as requiring that the polarizabilities of the TLS and the nanoparticle "overlap" each other, as shown in Figs. 3(a)–3(c). When such "overlapping" takes place, the conditions in Eq. (2) are satisfied when $r_0 \ll (\alpha_0)^{1/3}$ and $r_2 \ll (\alpha_2)^{1/3}$ [see Fig. 3(a)]. This corresponds, for example, to a metallic nanoparticle excited at the plasmon resonance frequency and a resonant atom or quantum dot with broadening close to the natural broadening limit. According to the results presented earlier, such a situation can be realized at low temperatures for the parameters assumed in Fig. 2. Higher-temperature operation requires a search for nanoparticles with a narrower plasmon resonance and/or TLS with higher polarizability.

The threshold condition of Eq. (15) cannot be satisfied if the polarizabilities of both particles are smaller then their volumes. However, condition (15) can be satisfied, in principle, if only one polarizability substantially exceeds the volume of the particle, for example, when $(\alpha_2)^{1/3} \gg r_2$, but $r_0 \leqslant (\alpha_0)^{1/3}$, as shown in Fig. 3(b). This implies that a plasmon resonance may not be strictly necessary in the nanoparticle

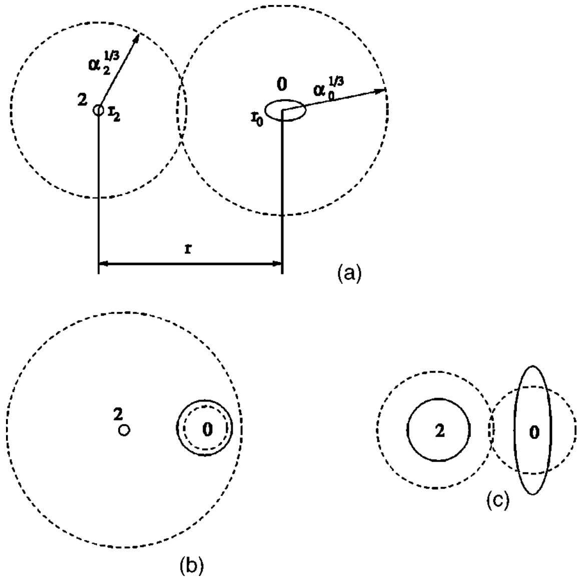

FIG. 3. Schematic of different approaches to achieve overlapping polarizabilities for dipole nanolaser; 2 indicates two-level system, 0 the nanoparticle. Dashed curves reflect the values of the polarizabilities, the solid curves show the geometrical size of the particles.

for dipole lasing, although it undoubtedly helps to satisfy the threshold condition for large values of interparticle separation. One could take a dielectric nanoparticle with $(\alpha_0)^{1/3} \sim r_0$, and the conditions given by Eq. (2) could still be satisfied. Finally, polarizability overlapping is possible even if both r_0 and r_2 are just a little smaller that $\alpha_0^{1/3}$ and $\alpha_2^{1/3}$ respectively, as shown in Fig. 3(c). For the cases shown in Figs. 3(b) and 3(c), however, Eq. (2) is no longer satisfied. It is then necessary to include quadrupole and possibly higher-order multipole moments in the interaction $\hat{V}$ between the TLS and the nanoparticle when estimating the DNL lasing threshold value. One can do this by utilizing the procedure of [17] to calculate the SP modes responsible for the nondipole interaction between the particles. Then one has to eliminate adiabatically the SP modes to derive the interaction term Ω_{int}.

In the meanwhile, we suggest that the requirement of "overlapping polarizability" provides the necessary condition for a dipole nanolaser, which remains useful when considering a general interaction $\hat{V}$ between the particles. The verification of this hypothesis will be done elsewhere in the frame of a more general model, beyond the dipole approximation. In support of this hypothesis, we note that the threshold condition for an ordinary laser is $2\pi\alpha_{2r}Q_0/V_m > 1$, which can be interpreted in the present context as requiring that the polarizability $2\pi\alpha_{2r}Q_0$ of an atom with respect to a cavity mode is larger than the cavity mode volume V_m.

An important condition for dipole lasing is that any anharmonicity in the polarization oscillations of the nanoparticle is small, so that the polarization of one particle has Bosonic properties. This condition can be broken when the nanoparticles become too small. As an "extreme" limit, for example, dipole lasing is not possible in the case of two atoms close to each other [29].

It order to make the analysis as straightforward as possible, we have presented here the simplest scheme for a dipole nanolaser. Other schemes can also be considered, including, for example, a dipole nanolaser array with more then one TLS or (and) nanoparticles. With the increase in the number of particles involved in lasing, the threshold conditions can be more easily satisfied and the lasing rate can be increased. One can also insert the dipole laser in a cavity, which reduces considerably the threshold conditions, but increases the size of the laser. Instead of a small number of particles, one may consider also the case of a continuous active medium with a large number of TLSs embedded, and with an ordered or disordered distribution of nanoparticles [13]. One can also construct layered structures, where an active semiconductor layer has a layer of nanoparticles above it [30]. A dipole nanolaser can act as an amplifier for external radiation, and also when the threshold conditions of Eq. (15) are not satisfied. Similar to the case of an ordinary laser with an external signal, the dipole nanolaser (nanoamplifier) can show bistable behavior in the presence of an external electromagnetic field. We suggest that a good starting point for experimental studies of dipole lasing is to search for a narrowing in the spectrum of the radiation scattered by a TLS with population inversion near or on the surface of a nanoparticle. Finally, we remark that dipole nanolasers can find applications in quantum optics as single-photon sources, a necessary prerequisite for many quantum optics experiments [31].

VII. CONCLUSION

We propose a "dipole nanolaser" (DNL) consising of a metallic nanoparticle coupled through a near field with a two-level system (TLS), with population inversion provided by an external pump. The transition frequency of TLS is close to the plasmon resonance frequency of the nanoparticle. We found threshold conditions, when nonzero dipole momenta (polarization) of TLS and the nanoparticle appears even at the absence of external electromagnetic field; we call this phenomenon "dipole lasing." Because of this nonzero polarization, TLS and nanoparticles emit coherent dipole radiation into free space. Dynamics of the polarization of particles obeys standard Maxwell-Bloch equations, where nanoparticle dipole moment stands for a lasing mode. This enables us to apply to DNL some well-known results of standard laser theory, in particular, the narrowing in the radiation spectrum with the increase in the pump above the dipole lasing threshold. Specific conditions necessary for dipole lasing is that the distance r between the nanoparticle and TLS must be smaller than the critical value r_{cr}; this condition guarantees strong coupling of particles through the near field. In the limit of the dipole-dipole interaction between particles $r < r_{cr}$ means that polarizabilities of TLS and the nanoparticle overlap in space. DNL does not need an optical cavity; this is why the typical size of DNL may be smaller than the lasing wavelength. As an example, DNL with a silver ellipsoidal nanoparticle coupled with a semiconductor quantum dot is considered. It is shown that such a DNL can operate, in principle, for typical parameters of TLS and silver nanoparticles; however, the narrowing in the plasmon resonance and TLS transition as, for instance, at low temperatures, help to satisfy threshold conditions. We compared DNL with an ordinary laser and with SPASER proposed in [17].

ACKNOWLEDGMENTS

I. E. Protsenko is grateful to the Russian Foundation for Basic Research, Grant No. 04-02-16211, A. V. Uskov is grateful for RFBR Grant No. 05-02-16537, for support. We also thank Science Foundation Ireland for financial support.

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