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Quantum plasmonics of metamaterials: loss compensation using spasers

A P Vinogradov, E S Andrianov, A A Pukhov,
 A V Dorofeenko, A A Lisyansky

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

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 quantum-plasmon device which consists of inversely excited two-level quantum dots (QDs) (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 $(kr_{\text{NP-TLS}})^{-3}$ times higher than the probability of radiative photon emission [15] ($r_{\text{NP-TLS}}$ 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

A P Vinogradov, E S Andrianov, A A Pukhov, A V Dorofeenko
 Institute for Theoretical and Applied Electromagnetics,
 Russian Academy of Sciences, Moscow, Russian Federation;
 E-mail: a-vinogr@yandex.ru

A A Lisyansky Department of Physics, Queens College
 of the City University of New York, USA

Uspekhi Fizicheskikh Nauk **182** (10) 1122–1130 (2012)
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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.

external field makes difficult the employment of spasers as nanodimensional devices, but these difficulties are not insurmountable. In particular, Stockman [12] came up with the idea that a spaser operating in the transient mode can be used as an amplifier. Therefore, spaser physics is interesting enough to be a research subject in its own right. Spaser physics constitutes a new area of optics—quantum plasmonics. For the development of metamaterial electrodynamics, however, of interest is the consideration of structures made up of ordered linear or two-dimensional spaser arrays rather than the treatment of a single spaser. In this case, collective interactions between spasers may significantly change the oscillation conditions and the properties of free-running spaser self-oscillations, and even give rise to new effects or instabilities in these structures. In this connection, along with plasmons localized on plasmon particles, of special interest are plasmons traveling along one-dimensional objects like wire, a chain of nanoparticles, or a groove in a metal [61–64]. The presence of an amplifying medium results in the amplification of one-dimensional plasmons [4, 23, 44, 65]. Although the presently existing theoretical estimates and experimental realizations in the area of quantum plasmonics rely on quite simple models, they show the promise of using spasers in the development of the elements of optical information facilities and optical computers.

In this report, we consider both individual and collective behavior of spasers in above-threshold oscillation.

2. Equations of spaser ‘motion’

Since the SP wavelength λ_{SP} is much shorter than the radiation wavelength λ in a vacuum [15, 16], the spatial derivatives in the Maxwell equations are much greater than the temporal ones. Neglecting the latter permits describing the plasmon field in the quasistatic approximation [66, 67]. It turned out that this is also true when the spaser operates even in the radiating nanoantenna mode, i.e., when the Joule loss in a nanoparticle is lower than the radiative loss. Considering the modes of a small spherical NP of radius $r_{\text{NP}} \ll \lambda$ shows the plasmon resonance frequency coincides with the frequency at which the NP is a half-wave antenna (resonator): a half of the plasmon wavelength fits into the sphere diameter [66].

Below, we shall consider the excitation of only the principal (dipole) SP mode with a frequency ω_{SP} . For a silver NP surrounded by silicon oxide (SiO_2), the permittivity values are well known [68]. Assuming that the NP radius $r_{\text{NP}} \sim r \sim 10$ nm, we estimate the dipole moment of the NP near the plasmon resonance: $\mu_{\text{NP}} \sim 200$ D. The dipole moment of a typical QD of size $r_{\text{TLS}} \sim 10$ nm is $\mu_{\text{TLS}} \approx 20$ D [69]. The NP–QD interaction adheres to a dipole–dipole one: $V = \hbar\Omega_{\text{R}} \sim \mu_{\text{NP}} \mu_{\text{TLS}}/r^3$, and the constant of this interaction (the Rabi frequency Ω_{R}) turns out to be two orders of magnitude lower than the oscillation frequency [22]. This permits us to apply the slowly varying amplitudes approximation below.

At the plasmon resonance frequency, the NP polarization is described by the oscillator equation with eigenfrequency equal to the plasmon resonance frequency:

$$\ddot{\mathbf{d}}_{\text{NP}} + \omega_{\text{SP}}^2 \mathbf{d}_{\text{NP}} = 0. \quad (1)$$

This oscillator is quantized in the standard way [59, 70]: the Bose operators are introduced in this case for the production $[\hat{a}^\dagger(t)]$ and annihilation $[\hat{a}(t)]$ of a dipole SP excited in the NP, which satisfy the commutation relation $[\hat{a}(t), \hat{a}^\dagger(t)] = 1$, and

the Hamiltonian is expressed as

$$\hat{H}_{\text{SP}} = \hbar\omega_{\text{SP}} \hat{a}^\dagger \hat{a}. \quad (2)$$

In the case of a spherical NP, the electric dipole mode field is uniform inside the NP, $\mathbf{E}_1 = -\boldsymbol{\mu}_1/r_{\text{NP}}^3$, and has the form $\mathbf{E}_1 = -\boldsymbol{\mu}_1/r^3 + 3(\boldsymbol{\mu}_1 \cdot \mathbf{r})\mathbf{r}/r^5$ outside. The vector of a unit dipole moment $\boldsymbol{\mu}_1$ is a dimensional quantity, and henceforward we shall explicitly write the factor $|\boldsymbol{\mu}_1|$.

The energy $\hbar\omega_{\text{SP}}$ of one plasmon is expressed as [71]

$$W_1 = \frac{1}{8\pi} \int_{V_{\text{NP}}} \omega \left. \frac{\partial \text{Re } \varepsilon}{\partial \omega} \right|_{\omega_{\text{SP}}} E_1 E_1^* dV_{\text{NP}} = \frac{|\boldsymbol{\mu}_1|^2}{6r_{\text{NP}}^3} \omega \left. \frac{\partial \text{Re } \varepsilon}{\partial \omega} \right|_{\omega_{\text{SP}}}, \quad (3)$$

where V_{NP} is the NP volume. Hence, the field produced by the NP can be written out as

$$\mathbf{E} = \sqrt{\frac{3\hbar r_{\text{NP}}^3}{|\boldsymbol{\mu}_1|^2 \partial \text{Re } \varepsilon / \partial \omega}} \mathbf{E}_1(\mathbf{r})(\hat{a} + \hat{a}^\dagger),$$

and accordingly the dipole moment of the NP is $\hat{\mathbf{d}}_{\text{NP}} = \boldsymbol{\mu}_{\text{NP}}(\hat{a} + \hat{a}^\dagger)$, where

$$\boldsymbol{\mu}_{\text{NP}} = \sqrt{\frac{3\hbar r_{\text{NP}}^3}{\partial \text{Re } \varepsilon_{\text{NP}} / \partial \omega}} \frac{\boldsymbol{\mu}_1}{|\boldsymbol{\mu}_1|^2}.$$

This is consistent [20, 21] with the ‘classical’ definition of the dipole moment [72]:

$$\mathbf{d}_{\text{NP}} = \frac{\varepsilon_{\text{NP}}(\omega) - \varepsilon_{\text{M}}}{\varepsilon_{\text{NP}}(\omega) + 2\varepsilon_{\text{M}}} \mathbf{E}_1 r_{\text{NP}}^3.$$

To describe the quantum dynamics of an NP and the two-level QD of a spaser, use can be made of a model Hamiltonian in the form [8, 54, 73]

$$\hat{H} = \hat{H}_{\text{SP}} + \hat{H}_{\text{TLS}} + \hat{V} + \hat{I}, \quad (4a)$$

where \hat{H}_{TLS} is the Hamiltonian of the two-level QD [16, 54, 74]:

$$\hat{H}_{\text{TLS}} = \hbar\omega_{\text{TLS}} \hat{\sigma}^\dagger \hat{\sigma}, \quad (4b)$$

operator $\hat{V} = -\hat{\mathbf{d}}_{\text{NP}} \hat{\mathbf{E}}_{\text{TLS}}$ defines the interaction between the two-level QD and the NP, and operator \hat{I} describes the relaxation and pumping effects [74]. The operator of the QD dipole moment is $\hat{\mathbf{d}}_{\text{TLS}} = \boldsymbol{\mu}_{\text{TLS}}(\hat{\sigma}(t) + \hat{\sigma}^\dagger(t))$, where $\hat{\sigma} = |g\rangle\langle e|$ is the transition operator between the excited $|e\rangle$ and ground $|g\rangle$ states of the QD, and $\boldsymbol{\mu}_{\text{TLS}} = \langle e|\hat{\mathbf{d}}_{\text{TLS}}|g\rangle$ is the transition dipole moment of the QD. Therefore, one obtains

$$\hat{V} = \hbar\Omega_{\text{R}}(\hat{a}^\dagger + \hat{a})(\hat{\sigma}^\dagger + \hat{\sigma}),$$

where the Rabi frequency

$$\Omega_{\text{R}} = \frac{\boldsymbol{\mu}_{\text{NP}} \boldsymbol{\mu}_{\text{TLS}} - 3(\boldsymbol{\mu}_{\text{TLS}} \mathbf{e}_r)(\boldsymbol{\mu}_{\text{NP}} \mathbf{e}_r)}{\hbar r^3},$$

and \mathbf{e}_r is a unit vector: $\mathbf{e}_r = \mathbf{r}/r$.

We assume that the QD transition frequency is close to the SP frequency, $\omega_{\text{SP}} \approx \omega_{\text{TLS}}$, and seek the solution in the form $\hat{a}(t) \equiv \hat{a}(t) \exp(-i\omega t)$ and $\hat{\sigma}(t) \equiv \hat{\sigma}(t) \exp(-i\omega t)$, where $\hat{a}(t)$ and $\hat{\sigma}(t)$ are slowly varying amplitudes. Then, neglecting rapidly oscillating terms $\sim \exp(\pm 2i\omega t)$ (the rotating wave

approximation [74]), the interaction operator \hat{V} can be written out in the form of the Jaynes–Cummings Hamiltonian [70]:

$$\hat{V} = \hbar\Omega_R(\hat{a}^\dagger\hat{\sigma} + \hat{\sigma}^\dagger\hat{a}). \quad (4c)$$

We proceed from Hamiltonian (4) and employ the standard commutation relations $[\hat{a}, \hat{a}^\dagger] = 1$, $[\hat{\sigma}^\dagger, \hat{\sigma}] = \hat{D}$ for operators $\hat{a}(t)$, $\hat{\sigma}(t)$ and the population inversion operator $\hat{D}(t)$ to arrive at the following Heisenberg equations of motion [8, 75]:

$$\dot{\hat{D}} = 2i\Omega_R(\hat{a}^\dagger\hat{\sigma} - \hat{\sigma}^\dagger\hat{a}) - \frac{\hat{D} - \hat{D}_0}{\tau_D}, \quad (5)$$

$$\dot{\hat{\sigma}} = \left(i\delta - \frac{1}{\tau_\sigma}\right)\hat{\sigma} + i\Omega_R\hat{a}\hat{D}, \quad (6)$$

$$\dot{\hat{a}} = \left(i\Delta - \frac{1}{\tau_a}\right)\hat{a} - i\Omega_R\hat{\sigma}, \quad (7)$$

where $\delta = \omega - \omega_{\text{TLS}}$ and $\Delta = \omega - \omega_{\text{SP}}$ are frequency mismatches. The QD population inversion operator $\hat{D}(t) = \hat{n}_e(t) - \hat{n}_g(t)$, where $\hat{n}_e = |e\rangle\langle e|$ and $\hat{n}_g = |g\rangle\langle g|$ are the operators of the upper and lower QD level populations, with $\hat{n}_e + \hat{n}_g = 1$. It should be emphasized that the population inversion operator $\hat{D}(t)$ is ‘slow’ on the strength of its definition. The contribution of relaxation and pumping effects, which is denoted by operator \hat{F} in Eqn (4a), is described in Eqns (5)–(7) by terms proportional to the relaxation rates τ_D^{-1} , τ_σ^{-1} , and τ_a^{-1} , and the operator \hat{D}_0 describes the population inversion produced by extraneous pumping in the QD [70, 74].

Strong dissipation in the NP makes this quantization scheme approximate and at the same time permits neglecting quantum correlations [8, 10]. This allows treating $\hat{D}(t)$, $\hat{\sigma}(t)$, and $\hat{a}(t)$ as complex quantities and replacing the Hermitian conjugation by the complex one [8, 10, 12, 43]. In this case, the quantity $D(t)$, which has the meaning of the difference between upper and lower level populations, will assume only real values, because the corresponding operator is Hermitian. The quantities $\sigma(t)$ and $a(t)$ have the meanings of dimensionless complex oscillation amplitudes of the dipole moments of the QD and the SP, respectively. Therefore, the spaser equations (5)–(7) in this approximation are single-mode optical Bloch equations [74].

3. Stationary spaser oscillation mode

Apart from the trivial solution $a = 0$, $\sigma = 0$, $D = D_0$ stable below the oscillation threshold, the system of equations (5)–(7) also has a nontrivial stationary solution:

$$a = \frac{\exp(i\varphi)}{2} \sqrt{\frac{(D_0 - D_{\text{th}})\tau_a}{\tau_D}}, \quad (8a)$$

$$\sigma = \frac{\exp(i\psi)}{2} \sqrt{\frac{(D_0 - D_{\text{th}})(\delta_{\text{SP}}^2 + \tau_a^{-2})\tau_a}{\Omega_R^2\tau_D}}, \quad (8b)$$

$$D = D_{\text{th}}, \quad (8c)$$

which corresponds to stationary spaser oscillation with a frequency $\omega = (\omega_{\text{SP}}\tau_a + \omega_{\text{TLS}}\tau_\sigma)/(\tau_a + \tau_\sigma)$, and the phases φ and ψ satisfy the relation

$$\cos(\psi - \varphi) = \frac{1}{\sqrt{1 + \tau_a^2(\delta - \Delta)^2}}.$$

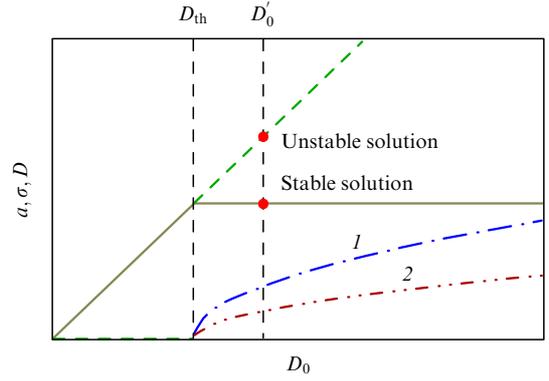


Figure 1. Stable stationary values of a (curve 1), σ (curve 2), and D (solid line) amplitudes. The unstable solution, which manifests itself for $D > D_{\text{th}}$, is shown by a dashed line. The stable and unstable solutions at some pumping rate $D_0 = D'_0$ are marked with black circles.

This solution is stable when the pumping D_0 exceeds the threshold value

$$D_{\text{th}} = \frac{1 + \Delta^2\tau_a^2}{\Omega_R^2\tau_a\tau_\sigma}. \quad (8d)$$

In this case, the stationary value of population inversion is fixed at the value of $D = D_{\text{th}}$ and ceases to increase with enhancing pumping (Fig. 1) [8, 12, 43].

4. Transition to stationary oscillations, Rabi oscillations

The use of spaser-based metamaterials implies that this medium will modify the electromagnetic wave propagating through the medium. However, one would think that the existence of an eigenfrequency and an oscillation amplitude in a spaser is an impediment to the employment of spasers as inclusions which actively interact with the outer wave [6]. The interaction efficiency can be improved by operating in a transient mode. M Stockman’s numerical experiments [12] suggest that a spaser exhibits complicated, strongly nonlinear dynamics during the transient regime. In this case, the spaser oscillation amplitude may be several times higher than the amplitude both of initial and of stationary spaser oscillations, i.e., the spaser can operate as an amplifier.

The transition of a spaser to stationary self-oscillations is defined by three characteristic times: the NP and QD polarization relaxation times τ_a and τ_σ , and the population inversion relaxation time τ_D . Due to high losses in metals, the time τ_a proves to be shortest. The experimental value is $\tau_a \sim (10^{-14} - 10^{-13})$ s [76], which coincides with the estimate obtained from classical electrodynamics [43]. Typical experimental values of remaining lifetimes are $\tau_\sigma = 10^{-11}$ s and $\tau_D = 10^{-13}$ s [77–79]. Therefore, for a metallic NP and a semiconductor QD, we obtain the following time scales: $\tau_a < \tau_D \ll \tau_\sigma$. The total self-oscillation settling time is defined by the longest time, $\sim \tau_\sigma$.

Numerical simulations have shown that the character of the transient process is heavily dependent on the initial value of the NP’s dipole moment amplitude $a(0)$. For an initially ‘cold’ spaser, $a(0) \ll 1$, and the electric field of the NP is weaker than the field of the QD. In this case, the energy goes from the QD to the NP, and the $a(t)$ value tends to the stationary value (8a). For a high initial amplitude $a(0) \gg 1$, which may be achieved under the excitation of the NP by a nanosecond optical pulse [6, 12], the transient process is more complicated and proceeds two stages [22]. During the first

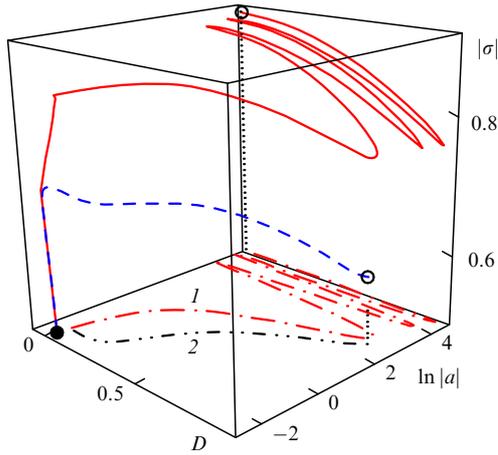


Figure 2. Spaser dynamics for $|\sigma| = 0.5$, $\tau_a = 10^{-14}$ s, $\tau_\sigma = 10^{-11}$ s, $\tau_D = 10^{-13}$ s, and $\Omega_R = 10^{13}$ s $^{-1}$. White dots correspond to the initial conditions for two trajectories emerging from $a(0) = 40 + 25i$, $\sigma(0) = 0.9$, $D(0) = 0.05$ (solid line) and from $a(0) = 5$, $\sigma(0) = 0.65$, $D(0) = 0.9$ (dashed line). The stationary state is indicated with a black dot. Curves 1 and 2 correspond to the projections of these trajectories onto the plane $|\sigma| = 0.5$.

stage, the QD passes time in the strong NP field and the spaser self-oscillation amplitude experiences Rabi oscillations with a characteristic period $\tau_R = 2\pi/\Omega_R$. As this takes place, the energy flux periodically changes its direction from the NP to the QD and vice versa. Over a time of order $\tau_a \ln|a(0)|$, these oscillations decay due to dissipation in the NP and the QD. During the second stage, the spaser exhibits dynamics characteristic for small $a(0)$, when the spaser oscillation amplitude tends monotonically to the stationary value. For the time relation $\tau_a < \tau_D \ll \tau_\sigma$ typical for the spaser, its dynamics depend only slightly on τ_D . The total duration of the transient mode is on the order of τ_σ (Fig. 2).

The frequency of oscillations observed in numerical simulations can be easily estimated in the case of exact resonance $\delta = \Delta = 0$. By omitting the terms responsible for relaxation and pumping, which may be done at the initial stage of the process when all spaser variables are distant from their stationary values, we arrive at the reduced system of equations

$$\dot{a} = -i\Omega_R \sigma, \quad (9)$$

$$\dot{\sigma} = i\Omega_R a D, \quad (10)$$

$$\dot{D} = 2i\Omega_R (a^* \sigma - \sigma^* a). \quad (11)$$

Substitution of Eqn (9) and its conjugate into Eqn (11) gives the Newton equation

$$\frac{d^2|a|}{dt^2} = \frac{\partial U(|a|)}{\partial a} \quad (12)$$

for a ‘particle’ of unit mass with coordinate $|a|$, which moves in the potential $U(|a|) = 0.5(\Omega_R^2|a|^4 - C_1\Omega_R^2|a|^2)$, where C_1 is the integration constant [22]. The stable equilibrium position for this ‘particle’ is $|a|_{\text{stable}} = \sqrt{C_1/2} = [|a(0)|^2 + D(0)/2]^{1/2}$, and the frequency of small oscillations about this equilibrium position is given by

$$\Omega = 2|a(0)|\Omega_R. \quad (13)$$

This expression coincides with the frequency of Rabi oscillations which emerge under the interaction of a two-level QD with a classical harmonic field of amplitude $a(0)$ or a quantized field with the number of photons approaching $\hat{a}^\dagger(0)\hat{a}(0) = n = |a(0)|^2$ [70].

5. Spaser in the field of an external optical wave, and spaser synchronization

Let us consider now the NP and QD dynamics in the field of an external optical wave $E(t) = E \cos(\nu t)$. Considering the external electric field as being classical and restricting ourselves to the dipole interaction, we write out the system’s Hamiltonian in the form

$$\begin{aligned} \hat{H}_{\text{eff}} = & \hat{H} + \hbar\Omega_1(\hat{a}^\dagger + \hat{a})[\exp(i\nu t) + \exp(-i\nu t)] \\ & + \hbar\Omega_2(\hat{\sigma}^\dagger + \hat{\sigma})[\exp(i\nu t) + \exp(-i\nu t)], \end{aligned} \quad (14)$$

where \hat{H} is defined by expression (4), and $\Omega_1 = -\mathbf{\mu}_{\text{NP}}\mathbf{E}/\hbar$ and $\Omega_2 = -\mathbf{\mu}_{\text{TLS}}\mathbf{E}/\hbar$ are the coupling constants of the NP and the QD to the external field.

As before, the equations of motion are the Heisenberg equations for the slowly varying amplitudes of operators \hat{a} , $\hat{\sigma}$, and \hat{D} :

$$\dot{\hat{D}} = 2i\Omega_R(\hat{a}^\dagger\hat{\sigma} - \hat{\sigma}^\dagger\hat{a}) + 2i\Omega_2(\hat{\sigma} - \hat{\sigma}^\dagger) - \frac{\hat{D} - \hat{D}_0}{\tau_D}, \quad (15)$$

$$\dot{\hat{\sigma}} = \left(i\delta_E - \frac{1}{\tau_\sigma}\right)\hat{\sigma} + i\Omega_R\hat{a}\hat{D} + i\Omega_2\hat{D}, \quad (16)$$

$$\dot{\hat{a}} = \left(i\Delta_E - \frac{1}{\tau_a}\right)\hat{a} - i\Omega_R\hat{\sigma} - i\Omega_1. \quad (17)$$

Here, $\delta_E = \nu - \omega_{\text{TLS}}$ and $\Delta_E = \nu - \omega_{\text{SP}}$ are frequency mismatches in the external optical field.

System of equations (15)–(17) has three stationary solutions $\{a_i, \sigma_i, D_i\}$, $i = 1, 2, 3$. A linear stability analysis of these solutions: $a(t) - a_i \sim \exp(\lambda t)$, $\sigma(t) - \sigma_i \sim \exp(\lambda t)$, and $D(t) - D_i \sim \exp(\lambda t)$ showed that only the solutions located in the lower branch of the curves depicted in Figs 3a and 3b are stable ($\text{Re } \lambda < 0$). For a zero mismatch $\Delta_E = \delta_E = 0$ in the absence of the field, the points indicated in Fig. 3a correspond to the points indicated in Fig. 1. For a nonzero mismatch, the stable solution branch $D(E)$ exists only when the field amplitude is sufficiently large: $E > E_{\text{synch}}(\Delta_E)$ (Fig. 3b).

Therefore, the value $E_{\text{synch}}(\Delta_E)$ is the lower boundary of the domain in which the spaser can be locked by an external wave. Such threshold behavior is typical for nonlinear systems experiencing external action, and the range of parameters E and Δ_E in which locking occurs is termed the Arnold tongue [80–82]. The boundary of the Arnold tongue can be qualitatively obtained by treating the external wave as a perturbation.

In the zero approximation in the field amplitude E , system (15)–(17) has the stationary solution (5)–(7). Let us find the solution in the first approximation in the field amplitude E . By substituting $a = |a| \exp(i\varphi)$ and $\sigma = |\sigma| \exp(i\psi)$ into Eqn (17), we obtain

$$\begin{aligned} \frac{d|a|}{dt} \exp(i\varphi) + i \frac{d\varphi}{dt} |a| \exp(i\varphi) \\ = \left(i\Delta_E - \frac{1}{\tau_a}\right) |a| \exp(i\varphi) - i\Omega_R |\sigma| \exp(i\psi) - i\Omega_1. \end{aligned} \quad (18)$$

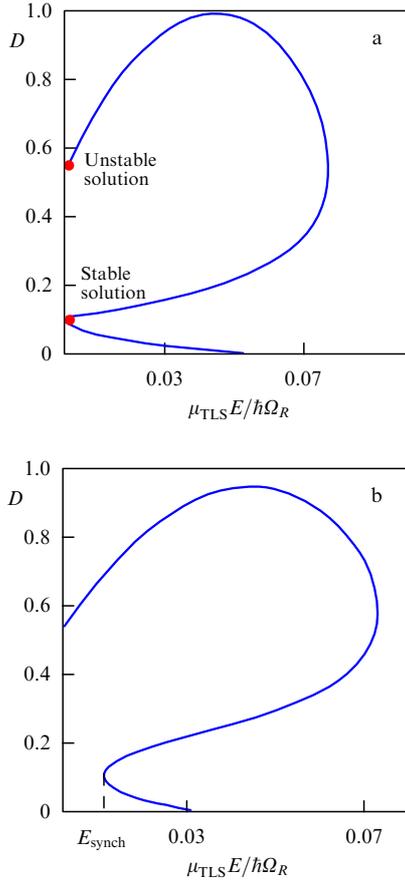


Figure 3. Stationary value of the population inversion D as a function of the external field amplitude for $\tau_a = 10^{-14}$ s, $\tau_\sigma = 10^{-11}$ s, $\tau_D = 0.5 \times 10^{-14}$ s, and $\Omega_R = 10^{13}$ s $^{-1}$: (a) $\Delta_E = \delta_E = 0$, and (b) $\Delta_E = \delta_E = 10^{11}$ s $^{-1}$.

We divide both sides of Eqn (18) by $|a| \exp(i\varphi)$ to bring the imaginary part of the equation to the form

$$\dot{\varphi} = \Delta_E - \Omega_R \frac{|\sigma|}{|a|} \cos(\psi - \varphi) - \frac{\Omega_1}{|a|} \cos \varphi. \quad (19)$$

Substituting the quantities (8) into Eqn (19) in place of $|a|$, $|\sigma|$, and $\cos(\psi - \varphi)$ yields the equations of motion [8, 22] of an overdamped ‘particle’ with a coordinate φ :

$$\dot{\varphi} = -\frac{\partial \Phi(\varphi)}{\partial \varphi} \quad (20)$$

in the potential $\Phi(\varphi) = -\Delta_E \varphi + \Omega_1 \sin \varphi / |a|$.

The phase dynamics are the sliding of this ‘particle’ over the potential profile in a viscous liquid. For $|\Omega_1| < |a\Delta_E|$, there occurs a unidirectional motion. The particle velocity oscillates with the period tending to infinity as the critical situation $|\Omega_1| = |a\Delta_E|$ is approached. For $|\Omega_1| > |a\Delta_E|$, the particle finds itself in one of the minima of the potential function $\Phi(\varphi)$, which corresponds to the synchronization mode: the oscillation phase φ is ‘locked’ and ceases to vary in time. Therefore, the spaser’s Arnold tongue is wedge-shaped in the low-field domain $E \ll \hbar\Omega_R / \mu_{\text{NP}}$.

Numerical simulations have shown that the boundary of the locking domain is described by the curve $E_{\text{synch}}(\Delta_E)$ (Fig. 4). Outside of this domain, the solution irregular in time, which corresponds to chaotic spaser behavior.

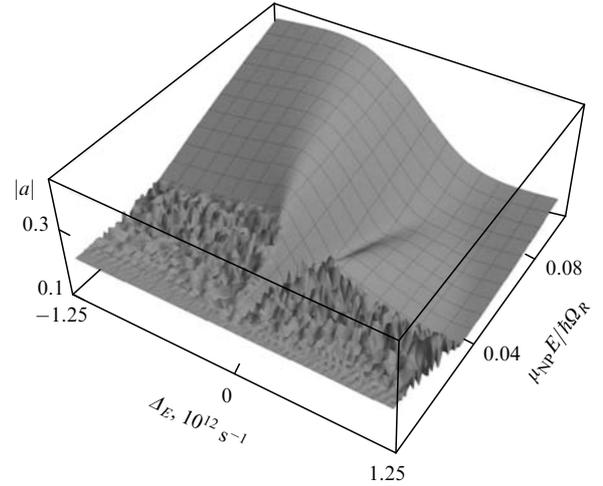


Figure 4. Dimensionless NP dipole moment as a function of external optical field amplitude E and frequency mismatch Δ_E . The speckle structure for small amplitudes of the external field corresponds to chaotic behavior of the dipole moment.

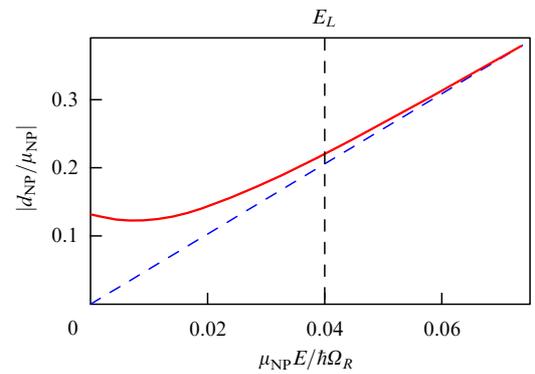


Figure 5. Amplitude of the NP dipole moment as a function of the magnitude of the external field for a zero mismatch.

Notice that a spaser which has reached the stationary state responds to the long-term ($\gg \tau_\sigma$) action of an external field in a qualitatively different manner than does a spaser exposed to a ‘pulsed’ ($\ll \tau_D$) external field, when the change in population inversion caused by the external field may be disregarded. While in the former case the response is nonlinear, in particular in a weak field $E \sim E_{\text{synch}}(\Delta_E)$ the dipole moment does not depend on the external field at all and is defined by the frequency mismatch and the pumping level (Fig. 5), in the latter case the spaser’s dipole moment is proportional to the external field [50, 56, 57].

In the absence of pumping ($D_0 = -1$), the solution of optical Bloch equations yields the answer close to the predictions of the classical theory describing the response of a single NP: the real part of an NP’s dipole moment can assume both positive and negative values, depending on the frequency, but its imaginary part is always positive. This corresponds to the energy transfer from the external field to the spaser. In the presence of a pump close in frequency to the spaser oscillation frequency, the imaginary part of the dipole moment may assume negative values for certain values of Δ_E , which corresponds to the energy transfer from the spaser to the external field (Fig. 6) [20].

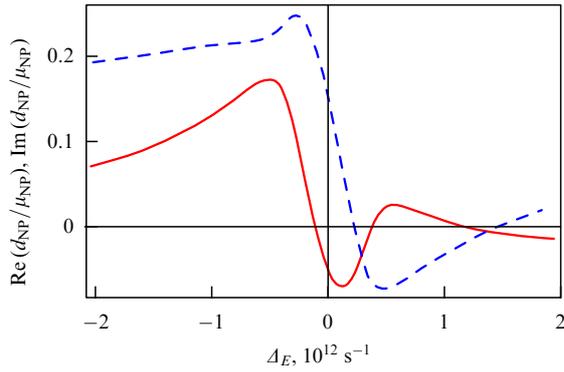


Figure 6. Real (solid curve) and imaginary (dashed curve) parts of the NP dipole moment as functions of frequency mismatch ΔE for an external field amplitude exceeding the locking threshold, $E > E_{\text{synch}}(\Delta E)$.

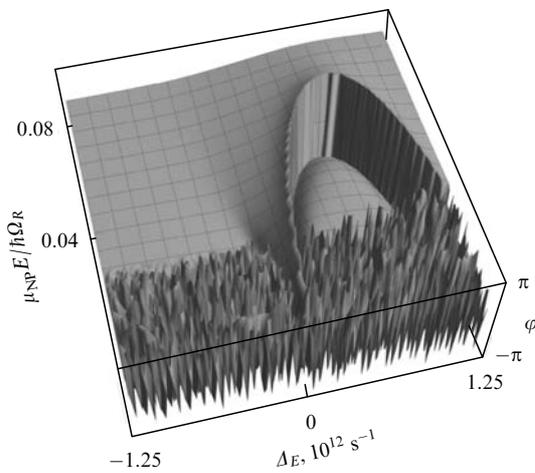


Figure 7. Dependence of $\varphi = \tan^{-1}(\text{Im } d_{\text{NP}}/\text{Re } d_{\text{NP}})$ on external field amplitude E and mismatch ΔE . The smooth part of the surface corresponds to the Arnold tongue region in which the spaser is synchronized by the external field. In the discontinuity line $E_{\text{com}}(\Delta E)$, where $\varphi = \pi$, the energy losses are precisely compensated.

It should be emphasized that for a given magnitude of external field there are two frequencies at which the imaginary part of the dipole moment turns to zero, i.e., complete compensation occurs (see Fig. 6). These points lie on the complete compensation curve $E = E_{\text{com}}(\Delta E)$ (Fig. 7), which is defined by the expression [20]

$$\left(\frac{\mu_{\text{NP}} E_{\text{com}}(\Delta E)}{\hbar}\right)^2 = \frac{1}{4} \left[-\frac{\tau_D \tau_\sigma^3}{\tau_a} \Delta E^4 + D_0 \Omega_R \tau_\sigma^3 \frac{\mu_{\text{TLS}}}{\mu_{\text{NP}}} \frac{\Delta E^3}{\tau_D} - \left(\frac{\tau_\sigma}{\tau_D} - \Omega_R^2 D_0 \frac{\tau_a \tau_\sigma^2}{\tau_D}\right) \Delta E^2 \right] \left(\tau_\sigma \Delta E \frac{\mu_{\text{TLS}}}{\mu_{\text{NP}}} + \tau_a \Omega_R \right)^{-2}. \quad (21)$$

As $\Delta E \rightarrow 0$, expression (21) changes into

$$\left(\frac{\mu_{\text{NP}} E}{\hbar}\right)^2 = (D_0 - D_{\text{th}}) \Delta E^2 \left(\frac{\tau_\sigma^2}{\tau_D \tau_a}\right).$$

Therefore, $E \propto (D_0 - D_{\text{th}})^{1/2} \Delta E$, and this curve lies inside of the Arnold tongue [20].

Figure 7 depicts the phase difference between the NP dipole moment and the external field, which was obtained by

the numerical solution of system (15)–(17). The discontinuity line, or the compensation line $E_{\text{com}}(\Delta E)$, corresponds to a phase difference π , when the imaginary part of the dipole moment is equal to zero. In this case, the real part of the NP dipole moment turns out to be negative.

If the external field amplitude corresponds to a point lying below the compensation curve, the energy is transferred from the spaser to the field, and as the wave field propagates over the system of spasers, its amplitude should increase, approaching the value $E_{\text{com}}(\Delta E)$. When the point lies above the curve, the energy will be absorbed inside the spaser, and the wave will attenuate and tend to the same value of amplitude. It is therefore expected that a wave will propagate over the system, whose amplitude will stably tend to the value defined by the level of spaser pumping and the frequency mismatch.²

The above reasoning relies on our analysis of the behavior of a single spaser. Moving from a single spaser to a spaser system may give rise to collective effects due to interspaser interaction, which may qualitatively change the picture of wave propagation through the active metamaterial.

6. Collective excitations of a spaser chain

So, we have ascertained that a spaser may synchronize its operation under the action of an external field. However, to make metamaterials requires the knowledge of how a spaser system works. In this case, collective interspaser interaction may significantly change the operating conditions and result in new effects. Indeed, since the times of Huygens it has been known that self-oscillating systems may synchronize their operation in the presence of even a weak interaction between them [81, 82]. Similar phenomena may also take place in a spaser system.

Below, we consider the collective interaction of spaser self-oscillations in the course of lasing above threshold by the simplest example of a linear spaser array. Two scenarios of the operation of the spaser system are possible in this case. First, the operation of all spasers may be synchronized, resulting in their in-phase operation. Second, a scenario is possible whereby the QD excitation will be transferred to their collective mode [2, 23, 65, 83, 84]. The role of collective mode is played by the dipole moment wave traveling along the chain of plasmon nanoparticles (see Ref. [60] and references cited therein).

For small frequency departures from the plasmon frequency, the dispersion relation for the wave of dipole moments traveling over an NP chain assumes the form

$$\omega(k) = \omega_{\text{SP}} + \gamma_i \frac{\omega_1^2}{\omega_{\text{SP}}} \cos(kb), \quad (22)$$

where $\omega_1^2 = r_{\text{NP}}^3 \omega_{\text{pl}}^2 / (3b^3)$, $\gamma_1 = 1$ for longitudinal modes, and $\gamma_1 = -2$ for transverse ones [60]. This solution becomes meaningless for $k < k_0 = \omega/c$, when the mode turns into a leaky one (see Ref. [85]), i.e., the radiation of photons occurs. For a single spaser, the radiationless excitation of plasmons prevails over the emission of photons for $(k_0 r_{\text{NP-TLS}})^3 \ll 1$, where $r_{\text{NP-TLS}}$ defines the characteristic scale of the system. In

² We emphasize: if a weak wave propagating over the system of spasers in the stochastic regime (outside of the Arnold tongue) were amplified, the system would be unstable. It would then be possible to observe spontaneous excitation of a wave with its amplitude defined by the ‘compensation line’.

the case of the collective mode, for a scale length r we must take k^{-1} , i.e., for $k < k_0$ we obtain $(k_0 r)^3 \geq 1$, and the energy will be transferred primarily to photons. Owing to the mutual synchronization of the spasers ($k = 0$), an effect similar to that considered in Ref. [14], where all spasers radiate in the same direction, should be observable in this situation.

The dipole–dipole interaction between the neighboring spasers gives rise to additional terms in Hamiltonian (4). Along with Ω_R , $\Omega_{\text{NP-NP}}$ —the coupling constant between nearest-neighbor NPs, $\Omega_{\text{NP-TLS}}$ —the coupling constant between the QD and the neighboring NPs, and $\Omega_{\text{TLS-TLS}}$ —the coupling constant between the nearest-neighbor QDs, appear.

The inclusion of $\Omega_{\text{NP-NP}}$ alone leads to the solution in the form of a harmonic wave with the dispersion equation

$$\omega_k = \omega_a + \Omega_{\text{NP-NP}}^{\text{eff}} \cos(kb),$$

where $\Omega_{\text{NP-NP}}^{\text{eff}} = 2\Omega_{\text{NP-NP}}\tau_a/(\tau_a + \tau_\sigma)$. This solution exists provided that the pumping exceeds the threshold value equal to

$$D_{\text{th}}(k) = \frac{1 + (\Omega_{\text{NP-NP}}^{\text{eff}}\tau_\sigma)^2 \cos^2(kb)}{\Omega_R^2 \tau_a \tau_\sigma}.$$

Despite the superficial similarity between the dispersion equations for the waves propagating over an NP chain and a spaser chain, there is a fundamental difference between these systems. First, the amplitude of waves propagating over the spaser chain is fixed and determined by the pumping level:

$$a_{n,k} = \frac{1}{2} \exp(i\varphi) \sqrt{\left[D_0 - \frac{1 + (\Omega_{\text{NP-NP}}^{\text{eff}}\tau_\sigma)^2 \cos^2(kb)}{\Omega_R^2 \tau_a \tau_\sigma} \right] \frac{\tau_a}{\tau_\sigma}}. \quad (23)$$

This autowave is different from solutions like solitons and kinks, which are known for other nonlinear systems [86], being a purely harmonic wave. Second, while the linear array of NPs obeys the superposition principle, an unusual situation takes place in the case of the chain of spasers: all autowaves, with the exception of the one with $k = \pm\pi/2b$, are unstable, and any initial perturbation evolves into this wave.

The inclusion of QD interaction with the neighboring NPs changes the situation qualitatively. The threshold pumping level becomes

$$D_{\text{th}}(k) = \frac{1 + (\tau_\sigma \Omega_{\text{NP-NP}}^{\text{eff}})^2 \cos^2(kb)}{[\Omega_R + 2\Omega_{\text{NP-TLS}} \cos(kb)]^2 \tau_a \tau_\sigma},$$

and the stability condition coincides with the condition for the minimum of $D_{\text{th}}(k)$ (Fig. 8).

One can see from Fig. 8 that a critical value exists for the coupling constant

$$\Omega_{\text{NP-TLS}}^* = \frac{1}{2} (\tau_\sigma \Omega_{\text{NP-NP}}^{\text{eff}})^2 \Omega_R$$

which separates frequency-dispersive waves from waves with $k = \pm\pi/b$ or $k = 0$ (Fig. 9). Waves with $k < k_0$ cannot travel along the linear chain, because they become leaky waves in this case [85].

7. Conclusion

There is a great demand for devices capable of manipulating light in domains smaller than the optical wavelength, i.e.,

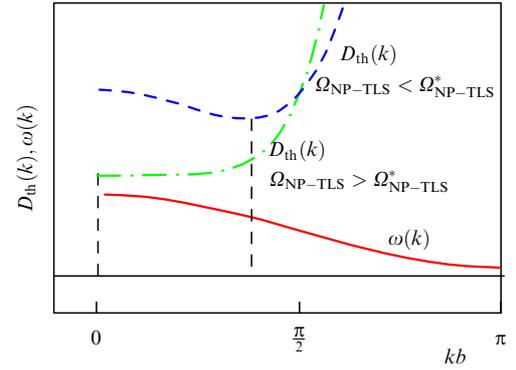


Figure 8. Dependence of the threshold pumping value on the wave vector k for $\Omega_{\text{NP-TLS}} < \Omega_{\text{NP-TLS}}^*$ (dashed curve) and $\Omega_{\text{NP-TLS}} > \Omega_{\text{NP-TLS}}^*$ (dot-and-dash curve). The solid curve depicts the $\omega(k)$ dependence.

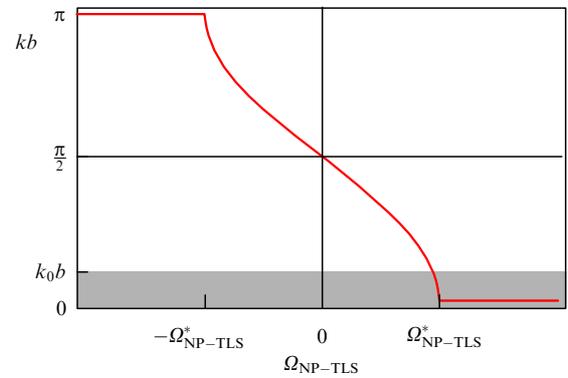


Figure 9. Dependence of the magnitude of a wave vector on the coupling constant between the neighboring NP and QD. The shaded domain corresponds to the leaky wave type solution.

measuring about ten nanometers: SNOM (scanning near-field microscopy), SERS, optoelectronic devices, etc. It is evident that coherent nanodimensional sources of optical radiation, i.e., nanolasers, rank with these devices. Among the possible ways of their realization is the spaser, with a plasmon nanoparticle fulfilling the function of a laser cavity.

However, it turns out that a spaser can be employed not only as a separate, ultrafast (with a response time of several femtoseconds) device, but also as an active inclusion in nanocomposites, including metamaterials. Indeed, a spaser-based composite material constitutes a new nonlinear object of research with unique properties. The spaser experiences Rabi oscillations in the transition to a stationary spasing mode, and therefore the properties of this material can be controlled by the intensity of external optical perturbation. In the stationary mode, the material reacts to the perturbation frequency by going from stochastic oscillations to the propagation of plasmon autowaves; the amplitude of the propagating waves depends only slightly on the amplitude of the incoming signal and is controlled primarily by the pumping level. These materials may show promise and find application in optoelectronics and metaplasmonics.

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