# Exactly solvable toy model for surface plasmon amplification by stimulated emission of radiation

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Abstract: We propose an exactly solvable electrodynamical model for surface plasmon amplification by stimulated emission of radiation (spaser). The gain medium is described in terms of the nonlinear permittivity with negative losses. The model demonstrates the main feature of a spaser: a self-oscillating state (spasing) arising without an external driving field if the pumping exceeds some threshold value. In addition, it properly describes synchronization of a spaser by an external field within the Arnold tongue and the possibility of compensating for Joule losses when the pumping is below threshold. The model also gives correct qualitative dependencies of spaser characteristics on pumping.

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OCIS codes: (240.6680) Surface plasmons; (350.4238) Nanophotonics and photonic crystals; (240.4350) Nonlinear optics at surfaces.

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#185509 - \$15.00 USD Received 19 Feb 2013; revised 4 Apr 2013; accepted 22 Apr 2013; published 25 Apr 2013 6 May 2013 | Vol. 21, No. 9 | DOI:10.1364/OE.21.010779 | OPTICS EXPRESS 10779 (C) 2013 OSA

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### 1. Introduction

Thanks to recent achievements of nanotechnology, quantum plasmonics has become one of the most rapidly growing areas of optics (see [1-6] and references therein). It is expected that nanoplasmonics will lead to a breakthrough in miniaturization of optical circuits, creating subwavelength optical components and on-chip integration of nanophotonics. The remarkable properties of nanoplasmonic devices are due to the ability of surface plasmons (SPs) to concentrate the optical energy at the space of much smaller size than the free space wavelength. However, the full development of nanoplasmonic devices is hindered by unacceptable level of Joule losses in metal nanostructures. The problem of Ohmic losses also slows down creation of numerous attractive applications based on metamaterials - man-made composites with negative dielectric permittivity and magnetic permeability (for review see [7,8]). To overcome this obstacle it was suggested introducing gain particles into plasmonic metallodielectric structures [1,3,5,9-17]. In these systems, a combination of metal nanoparticles (NP) with a nanoscale active medium having the population inversion results in the emergence of a nanoplasmonic counterpart of the laser – surface plasmon amplification by stimulated emission of radiation (spaser) first proposed by Bergman and Stockman [18] and considered as dipole nanolaser in [19]. This system was realized experimentally in [20–24].

The spaser is a quantum device aimed at enhancing the near field of SPs excited on a metal NP by a quantum system with population inversion, e.g. a quantum dot or dye molecules. Since our consideration is qualitative, below for simplicity we consider a generic two-level system (TLS). The TLS is excited by non-coherent pumping. Then it relaxes and excites SPs at the NP. The physical principle of spaser operation is similar to that of a laser. The role of photons confined to the Fabri-Perot resonator is played by SPs confineded to a NP [18,25-27]. Since the NP and the TLS are placed near to each other, the SPs excited on the NP trigger stimulated transition at the TLS, which in turn excites more SPs. This process is inhibited by losses in the NP, which together with pumping results in undamped stationary oscillations of the spaser dipole moment in the absence of external electromagnetic field (spasing) [6]. The spasing may occur only when the pumping exceeds some threshold value so that the energy supplied by pumping exceeds losses.

The main difference between spasers and lasers is that a spaser nonradiativly generates and amplifies the plasmonic mode localized on the NP in contrast to the travelling wave of a conventional laser. The SP amplification occurs due to radiationless energy transfer from the TLS to the NP. This process originates from the dipole-dipole (or any other near field [28]) interaction between the TLS and the plasmonic NP. This physical mechanism is highly efficient because the probability of SP excitation is approximately  $(kL)^{-3}$  times larger than the probability of radiative emission [29], where L is the distance between the centers of the NP and the TLS and k is the free space wavenumber.

The decrease of the system size leads to the increase of the ratio of the Joule losses to the rediation ones [30]. In fact, this ratio is determined by the same factor  $(kL)^{-3}$ . The size of the system considered in the present communication is chosen such that radiation losses can be disregarded in comparison to the Joule losses.

The spaser is a non-linear self-oscillating system, and therefore, its response to an external harmonic field principally differs from the response of isolated metal inclusions. Unlike an

isolated metal particle, the spaser can be synchronized with the external field only when the field amplitude exceeds a certain threshold value which depends upon the frequency of the field [16]. The domain of the field amplitude and frequency at which the synchronization occurs is called the Arnold tongue. Outside the Arnold tongue, the external field drives the spaser into stochastic oscillations. Inside the Arnold tongue, the energy supplied by pumping may compensate for Joule losses in the metal NPs [17] resulting in harmonic response of spaser on external field. Such compensation can be achieved even for values of pumping below the spasing threshold [31, 32].

Most of the results for spasers have been obtained in the framework of quantummechanical description (see, e.g., [6,12,14,16–19,27,32–35]). This description is reduced to the modified Maxwell-Bloch equations, in which quantum-mechanical operators are substituted by *c*-numbers. This approach is commonly used in the description of a single mode laser [36]. The same assumptions are made for the description of a gain medium in terms of permittivity with negative losses [37,38]. Thus, one can describe the TLS as a particle made of an amplifying medium, which has permittivity with negative losses. This approach is utilized in works [39–42], in which authors describe gain medium using a *linear* permittivity with a negative imaginary part. Although such an approach correctly demonstrates the lasing threshold of a nanolaser, it fails to reproduce nonlinear features of spasers, such as a stationary state with spasing, the spaser's behavior when pumping is above threshold, and the change in the population inversion by an external electromagnetic field. The authors of the cited articles realized the necessity of taking nonlinear effects into account, but attempts of doing so have a form of qualitative evaluations [43].

In this paper, we suggest an exactly solvable electrodynamical (more specifically, quasistatic) model of a spaser. The gain medium is assumed to be composed of dye molecules. The model is governed by the equations of classical electrodynamics and reflects the main features of spaser physics including the threshold transition to spasing. The quantum-mechanical effects are taking into account by introducing nonlinear permittivity. The model gives correct dependence of the SP amplitude on pumping and predicts existing of the region, in which a spaser may be synchronized by external wave, the Arnold tongue. The model also predicts compensation for losses by a spaser operating below the pumping threshold.

### 2. Theoretical model

## 2.1 Gain medium

The core of our consideration is the description of an amplifying (gain) medium in terms of dielectric permittivity. An expression for the permittivity may be deduced from the Maxwell-Bloch equations [37,38], which are commonly used in semiclassical descriptions of lasers [36,44] and spasers [19]. In the framework of this approach, the evolution of electric field  $\mathbf{E}$  is related to the macroscopic polarization  $\mathbf{P}$  of a gain subsystem via classical Maxwell's equation:

$$\nabla \times \nabla \times \mathbf{E} + \frac{\varepsilon_0}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2},\tag{1}$$

where  $\varepsilon_0$  is the dielectric constant of the host matrix and *c* is the speed of light in vacuum. Gain molecules embedded into the host medium are modeled as two-level systems with the transition dipole moment  $\mu$  spread in the host matrix. Dynamics of the polarization and the population inversion per unit volume *n* is governed by the equations following from the density matrix formalism [44]:

$$\frac{\partial^2 \mathbf{P}}{\partial t^2} + \frac{2}{\tau_p} \frac{\partial \mathbf{P}}{\partial t} + \omega_0^2 \mathbf{P} = -\frac{2\omega_0 |\boldsymbol{\mu}|^2}{\hbar} n\mathbf{E},$$
(2a)

$$\frac{\partial n}{\partial t} + \frac{1}{\tau_n} (n - n_0) = \frac{1}{\hbar \omega_0} \operatorname{Re} \left( \mathbf{E}^* \cdot \frac{\partial \mathbf{P}}{\partial t} \right), \tag{2b}$$

where  $\omega_0$  is the transition frequency of the two-level system,  $\tau_p$  and  $\tau_n$  are relaxation times for polarization and inversion, respectively, and  $n_0$  stands for pumping of active atoms. Assuming a harmonic time dependence of the electric field and the polarization and excluding the population inversion from this system we obtain the relation between the polarization and the electric field inside the medium, resulting in the following expression for nonlinear permittivity of a gain medium with an anti-Lorentzian profile [37,38]:

$$\varepsilon_{gain}(\omega) = \varepsilon_0 + D_0 \frac{\omega_0}{\omega} \frac{-i + (\omega^2 - \omega_0^2)/2\omega\Gamma}{1 + \beta |E(\omega)|^2 + [(\omega^2 - \omega_0^2)/2\omega\Gamma]^2},$$
(3)

where  $D_0 = 4\pi\mu^2 \tau_p n_0 / \hbar$  is a dimensionless parameter accounting for the stationary value of population inversion  $n_0$  of active atoms,  $\beta = \mu^2 \tau_n \tau_p / \hbar^2$ , and  $\Gamma = 1/\tau_p$  with typical values of polarization and population relaxation times for organic dyes being equal to  $\tau_n \sim 100 \text{ ps}$  and  $\tau_p \sim 10 \text{ fs}$ , respectively [45]. Below, we use the value of  $D_0$  as a characteristic of pumping of the gain medium.

In the current study, we focus on the interaction between lossy and gain media and their scattering properties rather than on the description of an amplifying medium. For this reason, we take some realistic values for  $\varepsilon_0$ ,  $\Gamma$ ,  $\omega_0$ , and  $D_0$ , and use them throughout the paper. Namely, we set  $\varepsilon_0 = 4$ ,  $\Gamma = 0.05 \,\text{eV}$ ,  $\omega_0 = 2 \,\text{eV}$ , and  $D_0 \le 0.25$ . The electric field is measured in units of  $\beta^{-1/2}$ . For our model, the estimated value of  $\beta^{-1/2}$  is  $\beta^{-1/2} = \hbar \sqrt{1/\tau_n \tau_p} / \mu \sim 10 \,\text{MV/m}$ .

## 2.2 Core-shell spaser

Consistent consideration of spaser of arbitrary design requires finding a solution of a nonlinear Laplace problem, which generally leads to very cumbersome calculations. As a model of a spaser we consider a spherical gain core of radius  $r_{core}$  coated with a silver plasmonic shell of radius  $r_{shell}$  (Fig. 1). This geometry allows us to exatly solve the nonlinear problem analytically. Indeed, in this system, the uniform field inside the core is a solution to the nonlinear Laplace equation. The radii used in our calculations are  $r_{shell} = 20 \text{ nm}$  and permittivity  $r_{core} = 0.85 r_{shell}$ . For silver the Drude formula we use  $\varepsilon(\omega) = \varepsilon_{\omega} - \omega_{pl}^2 / \omega / (\omega + i\gamma)$ . In order to fit actual experimental data [46], we use the parameters corresponding to silver:  $\varepsilon_{\infty} = 4.9$ ,  $\omega_{pl} = 9.5 \text{ eV}$ , and  $\gamma = 0.05 \text{ eV}$ . The plasmon resonance of a core-shell nanoparticle with such geometry splits into two resonances [47] having frequencies  $\omega_1 = 2.0 \text{ eV}$  and  $\omega_2 = 4.2 \text{ eV}$ . Recalling that the emission frequency of the gain medium is set to  $\omega_0 = 2 \,\text{eV}$ , one would anticipate SP amplification and spasing in the vicinity of frequency  $\omega_{\rm l}$ .



Fig. 1. A schematic drawing of a core-shell spaser.

Spasing is characterized by self-oscillation of spaser supported by pumping. For our model this corresponds to *nonzero dipole moment* of the core-shell NP at *zero applied external field*. To find this state we fix the potential  $\varphi_{core}(\mathbf{r}) = \mathbf{E}_{in} \cdot \mathbf{r}$  inside the gain core, which corresponds to the uniform field  $\mathbf{E}_{core}(\mathbf{r}) = -\nabla \varphi_{core} = \mathbf{E}_{in}$ . Note that in this case  $\Delta \varphi_{core} = 0$  even with nonlinear permittivity of the core. The potential inside the shell is chosen in the form  $\varphi_{shell} = -b\mathbf{E}_{in} \cdot \mathbf{r} + c\mathbf{E}_{in} \cdot \mathbf{r} / r^3$ , where *r* is the distance from the center of nanoparticle to the given point. It gives the field in the metallic shell as a linear response to the core field:

$$\mathbf{E}_{shell}(\mathbf{r}) = b\mathbf{E}_{in} - c\mathbf{E}_{in} / r^3 + 3c(\mathbf{E}_{in} \cdot \mathbf{n})\mathbf{n} / r^3, \qquad (4)$$

with  $\mathbf{E}_{shell}(\mathbf{r}) \cdot \mathbf{n} = b\mathbf{E}_{in} \cdot \mathbf{n} + 2c\mathbf{E}_{in} \cdot \mathbf{n} / r^3$  and  $\mathbf{E}_{shell}(\mathbf{r}) \times \mathbf{n} = b\mathbf{E}_{in} \times \mathbf{n} - c\mathbf{E}_{in} \times \mathbf{n} / r^3$ , where  $\mathbf{n} = \mathbf{r}/r$  is a unit vector. The potential outside the core-shell nanoparticle must vanish at the infinity and can be represented as  $\varphi_{out} = a\mathbf{E}_{in} \cdot \mathbf{r} / r^3$ . This gives the following expression for the field

$$\mathbf{E}_{out}\left(\mathbf{r}\right) = -a\mathbf{E}_{in} / r^3 + 3a(\mathbf{E}_{in} \cdot \mathbf{n})\mathbf{n} / r^3, \tag{5}$$

with  $\mathbf{E}_{out}(\mathbf{r}) \cdot \mathbf{n} = 2a\mathbf{E}_{in} \cdot \mathbf{n} / r^3$  and  $\mathbf{E}_{out}(\mathbf{r}) \times \mathbf{n} = -a\mathbf{E}_{in} \times \mathbf{n} / r^3$ . Note that the chosen form of the electrostatic potential  $\varphi_{out}$  corresponds to the electric dipole moment of the NP,  $\mathbf{d} = a\mathbf{E}_{in}$ . The three unknown coefficients, *a*, *b*, and *c*, can be found from the boundary conditions for the electromagnetic field at the inner and outer surfaces of the metallic shell [48]:

$$\begin{aligned} \mathbf{D}_{core} \cdot \mathbf{n} \Big|_{r_{core}} &= \mathbf{D}_{shell} \cdot \mathbf{n} \Big|_{r_{core}}, \quad \mathbf{E}_{core} \times \mathbf{n} \Big|_{r_{core}} &= \mathbf{E}_{shell} \times \mathbf{n} \Big|_{r_{core}}, \\ \mathbf{D}_{shell} \cdot \mathbf{n} \Big|_{r_{shell}} &= \mathbf{D}_{out} \cdot \mathbf{n} \Big|_{r_{shell}}, \quad \mathbf{E}_{shell} \times \mathbf{n} \Big|_{r_{shell}} &= \mathbf{E}_{out} \times \mathbf{n} \Big|_{r_{shell}}, \end{aligned}$$
(6)

where  $\mathbf{D} = \varepsilon \mathbf{E}$ . The condition of spasing corresponds to the non trivial solution for  $E_{in}$ . Substituting Eqs. (4) and (5) into the boundary conditions (6) and assuming non-zero value of  $E_{in}$ , we arrive at the following system of equations:

$$\begin{cases} \varepsilon_{gain} \left( \omega, E_{in}, D_0 \right) = \left( b + 2c / r_{core}^3 \right) \varepsilon_{shell}, \\ 1 = b - c / r_{core}^3, \\ \left( b + 2c / r_{shell}^3 \right) \varepsilon_{shell} = 2a / r_{shell}^3, \\ b - c / r_{shell}^3 = -a / r_{shell}^3. \end{cases}$$
(7)

Excluding unknown coefficients a, b, and c from system (7) we arrive at the following equation

$$\varepsilon_{gain}(\omega, E_{in}, D_0) = \varepsilon_{shell} \frac{2(\varepsilon_{shell}(\omega) - 1)r_{core}^3 - 2(2 + \varepsilon_{shell}(\omega))r_{shell}^3}{2(\varepsilon_{shell}(\omega) - 1)r_{core}^3 + (2 + \varepsilon_{shell}(\omega))r_{shell}^3}.$$
(8)

Since  $\varepsilon_{gain}$ , and  $\varepsilon_{shell}$  are complex values, Eq. (8) is a system of two equations for three real quantitities  $D_0$ ,  $E_{in}$ , and  $\omega$ . Thus, Eq. (8) implicitly defines the dependence  $E_{in}(D_0)$ .

## 3. Transition to spasing

Our immediate goal is finding the value of pumping,  $D_0$ , for which the field inside the core is not zero. Substituting Eq. (3) into Eq. (8) we obtain

$$\varepsilon_{0} + D_{0} \frac{\omega_{0}}{\omega} \frac{-i + \left(\omega^{2} - \omega_{0}^{2}\right)/2\omega\Gamma}{1 + \beta \left|E_{in}\right|^{2} + \left[\left(\omega^{2} - \omega_{0}^{2}\right)/2\omega\Gamma\right]^{2}} = F(\omega), \qquad (9)$$

where we introduce the notation

$$F(\omega) = \varepsilon_{shell} \frac{2(\varepsilon_{shell}(\omega) - 1)r_{core}^3 - 2(2 + \varepsilon_{shell}(\omega))r_{shell}^3}{2(\varepsilon_{shell}(\omega) - 1)r_{core}^3 + (2 + \varepsilon_{shell}(\omega))r_{shell}^3}.$$
 (10)

The left hand side of Eq. (9) depends on the frequency  $\omega$ , on the absolute value of the field inside the gain core,  $E_{in}$ , and on the gain  $D_0$ , while the right hand side,  $F(\omega)$ , depends on the frequency only. After some algebra one can see that Im  $F(\omega)$  is negative for a lossy shell. For any positive  $D_0$ , the imaginary part of  $\varepsilon_{gain}$  is negative as well and it is equal to zero for  $D_0 = 0$  (see Eq. (3)). Thus, there is always an interval,  $0 < D_0 < D_{th}$ , in which Im  $F(\omega) < \text{Im } \varepsilon_{gain}(\omega)$  so that Eq. (9) does not have solutions. The minimal value of the gain, for which Eq. (9) holds, can be considered as the threshold pumping for spasing. The corresponding frequency, at which Eq. (9) can be satisfied, is the *spasing frequency*,  $\omega_{sp}$ . Note that for  $D_0 = D_{th}$  and  $E_{in} = 0$ .

To find the value of  $\omega_{sp}$  we rewrite Eq. (9) as

$$\frac{D_0}{1+\beta |E_{in}|^2 + \left[\left(\omega^2 - \omega_0^2\right)/2\omega\Gamma\right]^2} = \frac{F(\omega) - \varepsilon_0}{-i + \left(\omega^2 - \omega_0^2\right)/2\omega\Gamma}\frac{\omega}{\omega_0}.$$
 (11)

Since the left hand side of this equation is real-valued, we obtain an equation that determines  $\omega_{sp}$ :

$$\operatorname{Im} \frac{F(\omega_{sp}) - \varepsilon_0}{-i + (\omega_{sp}^2 - \omega_0^2)/2\omega_{sp}\Gamma} = 0.$$
(12)

Equation (12) can be solved numerically to any desired accuracy. When Eq. (12) is satisfied, Eq. (11) is reduced to a single real-valued equation which determines the relation between the internal field  $E_{in}$  and  $D_0$ . Indeed, setting the frequency to  $\omega_{sn}$  in Eq. (11) we obtain:

$$\frac{D_0}{1+\beta \left|E_{in}\left(D_0\right)\right|^2 + \left[\left(\omega_{sp}^2 - \omega_0^2\right)/2\omega_{sp}\Gamma\right]^2} = \frac{\omega_{sp}}{\omega_0} \operatorname{Re} \frac{F(\omega_{sp}) - \varepsilon_0}{-i + \left(\omega_{sp}^2 - \omega_0^2\right)/2\omega_{sp}\Gamma}.$$
 (13)

In the previous paragraph we argued that  $E_{in} = 0$  and  $D_0 = D_{th}$  satisfy Eq. (9). An increase of  $D_0$  accompanied with an increase of  $E_{in}$  does not change the right hand side of Eq. (13). Thus,

$$\frac{D_0}{1+\beta |E_{in}(D_0)|^2 + \left[\left(\omega_{sp}^2 - \omega_0^2\right)/2\omega_{sp}\Gamma\right]^2} = \frac{D_{ih}}{1+0+\left[\left(\omega_{sp}^2 - \omega_0^2\right)/2\omega_{sp}\Gamma\right]^2}.$$
 (14)

Hence,  $E_{in}$ , as well as the dipole moment of the NP, depends on  $\sqrt{D_0 - D_{ih}}$ :

$$E_{in} = \sqrt{\left(D_0 - D_{ih}\right) / \beta D_{ih}} \sqrt{1 + \left[\left(\omega_{sp}^2 - \omega_0^2\right) / 2\omega_{sp}\Gamma\right]^2}, \qquad (15)$$
$$d = aE_{in}.$$

where  $a = \left[ -(1 + 2\varepsilon_{shell})(\varepsilon_{shell} - \varepsilon_{gain})r^3 + (\varepsilon_{shell} - 1)(2\varepsilon_{shell} + \varepsilon_{gain})R^3 \right] (9\varepsilon_{shell})^{-1}$ . This is similar to the bifurcation behavior in regular lasers and to that predicted for spasers [16,49]. Indeed, for  $D_0 = D_{th}$  the trivial stable point  $D = D_0$ , d = 0 becomes unstable and a new stable point D = $D_{th}$ ,  $d \neq 0$  arises with square-root dependence of d on  $(D_0 - D_{th})$ . Once  $\omega_{sp}$  is found numerically or analytically, one obtains threshold pumping from Eq.

(13):

$$D_{th} = \frac{\omega_{sp}}{\omega_0} \left( 1 + \left[ \left( \omega_{sp}^2 - \omega_0^2 \right) / 2\omega_{sp} \Gamma \right]^2 \right) \operatorname{Re} \frac{F(\omega_{sp}) - \varepsilon_0}{-i + \left( \omega_{sp}^2 - \omega_0^2 \right) / 2\omega_{sp} \Gamma}.$$
 (16)

Thus, our classical model predicts the threshold behavior for spasing, gives correct dependence of the amplitude of the spaser dipole oscillations on pumping, as well as expresses the values of threshold pumping and the intrinsic frequency of spasing via the parameters of the system. For the parameters specified above, the calculated spasing frequency and threshold are  $\omega_{sn} \approx 1.987 \text{ eV}$  and  $D_{th} \approx 0.25$ .

### 4. Loss compensation

As we mentioned in Introduction, including a gain medium into a metamaterial made of plasmonic NPs turns metamaterial into a matrix filled with spasers which inject energy into the system in order to compensate for loss. The general goal of such compensation is to construct a lossless metamaterial in which the electromagnetic response mimics the response of an ordinary composite without loss. Such metamaterial should be characterized by an effective dielectric permittivity. Such a description implies that the spaser response should monotonically depend on the amplitude of the external field and should oscillate with the frequency of the driving field. When the exact loss compensation occurs, the imaginary part of the spaser dipole moment is equal to zero [17].

In works [14,31,50] the possibility of loss compensation was studied by computer simulation. In order to be able to use the effective permittivity to describe the spaser response, the authors considered very short pulses of the external wave. It was assumed that during the pulse, the population inversion does not change. Thus, despite using nonlinear equations, the authors obtain results of the linear theory.

The present toy model of the spaser allows us to consider nonlinear response of the spaser. Above pumping threshold, a spaser is a self-oscillating system with fixed frequency and amplitude. Therefore, in this regime spasers are not very convenient for loss compensation for a wide range of frequencies [51,52]. Even though such a spaser can be synchronized by external optical field, so that it oscillates with the frequency of that field [16] and losses can be compensated at certain frequencies and amplitudes of that external field [17], the amplitude of spaser dipole oscillations weakly depends on the amplitude of the external field. The value of this amplitude is about the same as the amplitude of oscillations of a non-driven spaser.

The response of a spaser operating below pumping threshold is more suitable for compensating losses in a metamaterial matrix. Indeed, below pumping threshold a spaser does not oscillate without an external field. Such a spaser is always synchronized by the external

Received 19 Feb 2013; revised 4 Apr 2013; accepted 22 Apr 2013; published 25 Apr 2013 #185509 - \$15.00 USD 6 May 2013 | Vol. 21, No. 9 | DOI:10.1364/OE.21.010779 | OPTICS EXPRESS 10786 (C) 2013 OSA

field. The amplitude of the dipole oscillations nonlinearly depends on the strength of the external field. The question remains whether the driven below-threshold spaser could actually compensate for losses. The quantum-mechanical consideration of this problem is done in [32].

In order to calculate the response of a driven below-threshold spaser on the external incident field, we should include a respective term  $\mathbf{E}_{ext} = x\mathbf{E}_{in}$  into system of Eqs. (7). Assuming that the NP dipole moment oscillates with the frequency of the external field  $\omega$ , we obtain:

$$\begin{cases} \varepsilon_{gain} \left( \omega, E_{in}, D_0 \right) = \left( b + 2c / r_{core}^3 \right) \varepsilon_{shell}, \\ 1 = b - c / r_{core}^3, \\ \left( b + 2c / r_{shell}^3 \right) \varepsilon_{shell} = x + 2a / r_{shell}^3, \\ b - c / r_{shell}^3 = x - a / r_{shell}^3. \end{cases}$$
(17)

Numerical solution of Eqs. (17) gives the values of the NP dipole moment  $\mathbf{d} = a\mathbf{E}_{in}$  for a given frequency and the amplitude of the external field. Indeed, once frequency and internal field  $E_{in}$  are fixed, from system (17) we obtain all the unknown coefficients, in particular, *a* and *x* which define the dipole moment **d** and the incident field  $\mathbf{E}_{ext}$ , respectively.

and x which define the dipole moment **u** and the incident field  $\mathbf{E}_{ext}$ , respectively.

In Fig. 2 we show parametric plots of the dipole moment of the driven spaser versus external field for two regimes: below- and above-threshold spaser operation and for two different frequencies  $\omega_{\leq} = 1.98 \,\text{eV} < \omega_{sp}$  and  $\omega_{\geq} = 1.99 \,\text{eV} > \omega_{sp}$ . One can see that below the threshold, there is a range of external field amplitudes in which the imaginary part of the total dipole moment is negative (see Fig. 2(b)). In this regime, with the help of spasers, energy of pumping is transferred to the system and exceeds losses. For a certain amplitude, the imaginary part of dipole moment turnes to zero. At this point, losses in the composite NP are exactly compensated by energy transferred to the system by the spaser operating below spasing threshold. The external field amplitude for which exact loss compensation occurs depends upon the field frequency forming a curve of the exact compensation. As one can see from Fig. 2(a), there is no loss compensation for frequencies smaller that the spasing frequency  $\omega_{sn}$ . This agrees with the results of the quantum-mechanical approach [32], which shows that loss compensation at frequencies  $\omega < \omega_{sp}$  is impossible. As one can see from Fig. 2(d), the field required for loss compensation can be estimated as  $0.005\beta^{-1/2}$ , so that the typical loss compensation field is of the order of  $\sim 50$  kV/m which is much smaller than the breakdown field of a dielectric ( $\sim 10 \div 100 \text{ MV/m}$ ).

Our model shows that loss compensation can be achieved in the regime of above-threshold pumping as well (marked by a green circle in Fig. 2(d)). This is also in agreement with the quantum-mechanical consideration [16,17,53]. However, as we discuss above, in this regime the spaser's response to the external field is strongly non-linear which makes it less attractive for applications. Finally, as one can see from Fig. 2(c), above the threshold loss compensations may *formally* occur in the region of frequencies  $\omega < \omega_{sp}$  (marked by a red circle in Fig. 2(c)). However, as shown in the following section, this solution is unstable.

Recently, Khurgin and Sun (KS) [54,55] raised concerns of possibility of loss compensation by spasers and their practical realization. As KS discussed, in the injection pumped spasers, the threshold current density may reach unsustainable values. Here, we would like to emphasize the difference between our model and the one considered by KS. In contrast to papers [54,55], in which semiconductor optical devices with current pumping are studied, we consider organic dye which is pumped optically. Semiconductor and dye-based lasers belong to different classes of lasers [56]. These classes are characterized by relations between relaxation times. The population inversion relaxation time of organic dye is of the order of  $\tau_n \sim 10^{-10}$  s. Thus, the Purcell factor in our case is significantly smaller than one

 <sup>#185509 - \$15.00</sup> USD
 Received 19 Feb 2013; revised 4 Apr 2013; accepted 22 Apr 2013; published 25 Apr 2013

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 6 May 2013 | Vol. 21, No. 9 | DOI:10.1364/OE.21.010779 | OPTICS EXPRESS 10787

considered in Refs [54,55]. For typical relaxation times of the plasmon mode,  $\tau_a \sim 10^{-14}$  s, the polarization,  $\tau_p \sim 10^{-14}$  s, and the Rabi frequency,  $\Omega_R \sim 10^{13}$  s<sup>-1</sup>, the Purcell factor is of the order of  $1 + \Omega_R^2 \tau_a \tau_p \sim 1$ . This is by two orders of magnitude smaller than the evaluation obtained by KS. Moreover, KS studied the regime of developed spasing. This implies that the mean number of plasmons is greater than unity ( $n_{pl} \gg 1$ ). In this paper, we consider a spaser

operating near or below threshold. In this case [34],  $n_{pl} \le 0.1$  and local fields and pumping energy have reasonable values.



Fig. 2. Spaser response on external harmonic field in (top panels) below- and (bottom panels) above-threshold regimes. Solid and dashed lines show real and imaginary parts of the dipole moment, respectively. Frequencies of incident field are (a), (c)  $\omega_{<} = 1.98 \text{ eV} < \omega_{sp}$  and (b), (d)  $\omega_{>} = 1.99 \text{ eV} < \omega_{sp}$ . The red and green circles in the bottom panels show the exact loss compensation when the imaginary part of the dipole moment turns to zero.

The pump intensity can be estimated as the net power dissipated in the active medium and the plasmonic NP near threshold:  $W_{pump} \sim \hbar \omega_0 n_e N / \tau_n + \hbar \omega_{sp} n_{pl} / \tau_a$ , where  $n_e$  is the population of the excited state of the gain medium at threshold, which is of the order of 1. Therefore, for core radius of ~20 nm and gain molecules density of  $\rho \sim 10^{19}$  cm<sup>-3</sup> we obtain that inside the active core of spaser there are approximately  $N \sim r_{core}^3 \rho \sim 100$  of dye molecules. For value of  $\tau_n = 10^{-10} s$ , this yields the pump power  $W_{pump} \sim 10^{-5}$  W. The energy flux density is then given by  $W_{pump} / \lambda^2$ , where  $\lambda$  accounts for the resonant cross-section of the spaser interaction with the pumping optical wave (note that the interaction cross-section is not the squared spaser's radius,  $r_{core}^2$ , but the squared wavelength,  $\lambda^2$ ). Since the Pointing

vector of a plane wave is  $S = c^2 \varepsilon_0 |E|^2$  (where  $\varepsilon_0$  is the electric constant), we find an estimation for the electric field strength for the pumping wave  $E_{pump} \sim \sqrt{W_{pump} / (\varepsilon_0 c^2 \lambda^2)} \sim 10 \text{ V/m}.$ 

# 5. Spaser synchronization and the Arnold tongue

Above threshold spaser is a self-oscillating system. As it is known from the general theory of nonlinear oscillators [57], response of a self-oscillating system on an external harmonic perturbation may be either periodic or stochastic, i.e. oscillations of above-threshold spaser in an external optical field may be unstable. To shed light on this issue, we consider equations describing the temporal evolution of the dipole moment of a spaser driven by the external oscillating field.

To investigate the time evolution of the stationary state we cannot consider the external wave as a plane wave with a constant amplitude, but should consider a slowly varying long pulse of the external field  $\mathbf{E}_{ext}(t) = \mathbf{E}_{slow}(t) \exp(-i\omega t)$  and corresponding dipole moment,  $\mathbf{d}(t) = \mathbf{d}_{slow}(t) \exp(-i\omega t)$ , induced in a core-shell spaser. Here,  $\mathbf{d}_{slow}(t)$  and  $\mathbf{E}_{slow}(t)$  are slowly varying envelopes, which Fourier transformations include only frequencies that are much smaller than the central frequency  $\omega$ . The external field and dipole moment are related via the nonlinear operator,

$$\hat{\boldsymbol{\alpha}}^{-1}(t, E_{in}(t))\mathbf{d}(t) = \mathbf{E}_{ext}(t).$$
(18)

The explicit form of the operator  $\hat{\alpha}^{-1}(E_{in})$  is not known because it depends on the field inside the gain medium,  $E_{in}(t)$ , which is a long pulse as well, whereas permittivity (3) is obtained for a harmonic field with the amplitude  $E(\omega)$ . To make Eqs. (18) and (3) consistent with each other, we define the operator  $\hat{\alpha}^{-1}(E_{in})$  through its action on the harmonic field as follows:

$$\boldsymbol{\alpha}^{-1}(\boldsymbol{\omega}, E_{in}(\boldsymbol{\omega}))\tilde{\mathbf{d}}(\boldsymbol{\omega}) = \tilde{\mathbf{E}}_{ext}(\boldsymbol{\omega}), \qquad (19)$$

where  $E_{in}(\omega)$  is the value that should be put into Eq. (3) and '~' indicates the Fourier transformation of the corresponding quantity. In this case  $\alpha^{-1}$  is simply the relation between incident field and dipole moment and can be obtained from the system (17):  $\alpha^{-1} = E_{ext} / d = x / a$ .

Applying the Fourier transformation to Eq. (19) we get:

$$\mathbf{E}_{ext}(t) = \int \alpha^{-1}(\Omega, E_{in}) \tilde{\mathbf{d}}(\Omega) e^{-i\Omega t} d\Omega = e^{-i\omega t} \int \alpha^{-1}(\omega + \nu, E_{in}) \tilde{\mathbf{d}}_{slow}(\nu) e^{-i\nu t} d\nu.$$
(20)

Since the main contribution to integral (20) is given by harmonics having  $v \ll \omega$ , we obtain

$$\mathbf{E}_{ext}(t) \approx e^{-i\omega t} \int \left[ \alpha^{-1}(\omega, E_{in}) + \nu \frac{d\alpha^{-1}(\omega, E_{in})}{d\omega} \right] \tilde{\mathbf{d}}_{slow}(\nu) e^{-i\nu t} d\nu = \\ = e^{-i\omega t} \alpha^{-1}(\omega, E_{in}) \int \tilde{\mathbf{d}}_{slow}(\nu) e^{-i\nu t} d\nu + e^{-i\omega_{0}t} \frac{d\alpha^{-1}(\omega, E_{in})}{d\omega} \int \nu \tilde{\mathbf{d}}_{slow}(\nu) e^{-i\nu t} d\nu$$
(21)
$$= e^{-i\omega t} \left[ \alpha^{-1}(\omega, E_{in}) \mathbf{d}_{slow}(t) + i \frac{d\alpha^{-1}(\omega, E_{in})}{d\omega} \frac{d\mathbf{d}_{slow}(t)}{dt} \right],$$

where the term with d/dt accounts for small broadening of dipole moment spectra (see also [58,59]). Cancelling the oscillating factor  $e^{-i\alpha t}$  at both sides of Eq. (21) we arrive at the

desired equation describing temporal evolution of the spaser dipole moment in slowly varying external field with the central frequency  $\omega$ :

$$i\frac{d\alpha^{-1}}{d\omega}\frac{d}{dt}\mathbf{d}_{slow}(t) + \alpha^{-1}\mathbf{d}_{slow}(t) = \mathbf{E}_{slow}(t).$$
(22)

Provided  $\mathbf{E}_{slow}(t) \equiv \mathbf{E}$  is constant, Eq. (22) has a stationary solution  $\mathbf{d}_{slow} = \alpha \mathbf{E}$ . In order to study how small perturbations of this solution evolve with time, let us consider a perturbation in the form  $\mathbf{d}_{slow} = \alpha \mathbf{E} + \delta \mathbf{d} e^{\Delta t}$ . Substituting this into Eq. (22) we arrive at

$$i\frac{d\alpha^{-1}}{d\omega}\delta\mathbf{d}\Lambda + \alpha^{-1}\delta\mathbf{d} = 0.$$
 (23)

The instability growth rate  $\Lambda$  is then given by

$$\Lambda = i\alpha^{-1}(\omega, E_{in}) \left( d\alpha^{-1}(\omega, E_{in}) / d\omega \right)^{-1}.$$
(24)

The stationary solution of Eq. (22) becomes unstable when Re  $\Lambda > 0$ . In Fig. 3 we plot the real part of the instability growth rate for both below- and above-threshold regimes of a driven core-shell spaser for the incident field frequency  $\omega_c = 1.98 \text{ eV}$ .



Fig. 3. Real parts of the instability growth rate  $\Lambda$  for below- (solid line) and above-threshold (dashed line) regimes of a driven spaser at the frequency of incident field  $\omega_{<} = 1.98$  eV. For below-threshold spaser the instability growth rate is always negative so that spaser oscillations in external field are always stable. Above threshold, spaser is synchronized by strong external field, while its oscillations in weak external field are unstable and may show stochastic dynamics [16].

In the below-threshold regime, ReA is negative for any field. Thus, the steady-state oscillations are stable with respect to small perturbations. When pumping increases, so that  $D_0$  exceeds  $D_{th}$ , a region in which ReA > 0 arises. In this region, the steady-state solution becomes unstable. In the above-threshold regime without external field, the spaser oscillates with its spasing frequency, which is obtained from Eq. (12). When the external field is applied, depending on its amplitude and frequency, the spaser may or may not oscillate with the frequency of this field. When it has the same frequency as the external field, the steady-state solution is stable, and spaser is synchronized with the external field. The region in which the synchronization takes place is known the Arnold tongue [60]. This region is shown in Fig. 4 for  $D_0 = 0.25$ . When the frequency of the external field is tuned to the spasing frequency,  $\omega_{sp}$ , the synchronization occurs for vanishingly small amplitude of the external field.

# 6. Discussion

The model presented here reproduces general features of the quantum description of a spaser including the pumping threshold for spasing, the bifurcation behavior at the transition point, correct dependencies of spaser characteristics on pumping, and the existence of a region of

spaser synchronization with an external field – the Arnold tongue. Our semiclassical model also predicts the possibility of loss compensation by a spaser operating below threshold.



Fig. 4. Stable (filled) and unstable regions of a driven above-threshold spaser for  $D_0 = 0.25$ . The region of stability corresponds to steady-state solutions with  $\operatorname{Re} \Lambda < 0$ . The two vertical lines correspond to the frequencies  $\omega_{>}$  and  $\omega_{<}$ , which are also used in Fig. 2. The red and green circles correspond to those in Fig. 2 (c) and 2(d). The red circle, in which  $\operatorname{Im}[d] = 0$ , lies in the instability region, so loss compensation does not occur, while the green circle lies in the stable region and compensation does take place. The red and green curves show the compensation curves lying in unstable and stable regions, respectively, along which the imaginary part of dipole moment turns to zero.

Our model also reveals inconsistencies in linear models of nanolasers. In particular, the authors of [39] consider a metal-coated nanolaser and report a lasing turn-off above the threshold. This result is in disagreement with the experimental observation of spasing in core-shell nanolasers and with general theory of lasers [44]. The authors make a suggestion that the on/off behavior of lasing in coated nanoparticles is caused by detuning of the resonance when the gain is added. However, in our study we show that the spasing frequency does not depend on gain and is a function of nanolaser geometry only (see Eq. (12)).

Using our model, it is interesting to look at the discussion concerning the possibility of loss compensation in plasmonic systems with gain (see [52] and comments to this work). In paper [52], Stockman argues that in a resonant plasmonic structure Ohmic losses are compensated for by gain when spasing occurs. Indeed, this argument is valid for a closed system in which there is no incoming and outgoing radiation. In this case, loss compensation and lasing simply coincide.

In an open system coupled with the radiation, it is necessary to compensate for both Ohmic and radiation losses for spasing to occur. In this case, as we show above for a spaser operating below threshold, lossless scattering of an incoming wave may occur when the system does not spase. This happens because the magnitude of dipole oscillations is smaller than that in the above-threshold spaser and the pumping energy is, therefore, sufficient to compensate for the loss. This situation is analogous to the scheme suggested in [9], in which Ohmic losses in the illuminated photonic crystal composed of alternating metallic and dielectric amplifying layers are compensated below the lasing threshold.

#### Acknowledgments

The authors would like to thank A. A. Pukhov for helpful discussion. They also are indebted to R. E. Noskov for helpful discussion of Eq. (22). This work was supported by RFBR Grants Nos. 12-02-01093, 13-02-00407 by a PSC-CUNY grant and by the Dynasty Foundation.