

# Forced synchronization of spaser by an external optical wave

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**Abstract:** We demonstrate that when the frequency of the external field differs from the lasing frequency of an autonomous spaser, the spaser exhibits stochastic oscillations at low field intensity. The plasmon oscillations lock to the frequency of the external field only when the field amplitude exceeds a threshold value. We find a region of values of the external field amplitude and the frequency detuning (the Arnold tongue) for which the spaser synchronizes with the external wave.

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## References and links

1. W. Cai and V. Shalaev, *Optical Metamaterials* (Springer, 2010).
2. S. A. Ramakrishna and J. B. Pendry, "Removal of absorption and increase in resolution in a near-field lens via optical gain," *Phys. Rev. B* **67**(20), 201101 (2003).
3. A. Fang, T. Koschny, and C. M. Soukoulis, "Lasing in metamaterial nanostructures," *J. Opt.* **12**(2), 024013 (2010).
4. M. A. Noginov, G. Zhu, M. Bahoura, J. Adegoke, C. E. Small, B. A. Ritzo, V. P. Drachev, and V. M. Shalaev, "Enhancement of surface plasmons in an Ag aggregate by optical gain in a dielectric medium," *Opt. Lett.* **31**(20), 3022–3024 (2006).
5. A. K. Popov and V. M. Shalaev, "Compensating losses in negative-index metamaterials by optical parametric amplification," *Opt. Lett.* **31**(14), 2169–2171 (2006).
6. A. K. Sarychev and G. Tartakovsky, "Magnetic plasmonic metamaterials in actively pumped host medium and plasmonic nanolaser," *Phys. Rev. B* **75**(8), 085436 (2007).
7. M. A. Noginov, G. Zhu, V. P. Drachev, and V. M. Shalaev, "Surface plasmons and gain media," in *Nanophotonics with Surface Plasmons*, V. M. Shalaev and S. Kawata, eds. (Elsevier, 2007).
8. N. I. Zheludev, S. L. Prosvirnin, N. Papasimakis, and V. A. Fedotov, "Lasing spaser," *Nat. Photonics* **2**(6), 351–354 (2008).
9. E. Plum, V. A. Fedotov, P. Kuo, D. P. Tsai, and N. I. Zheludev, "Towards the lasing spaser: controlling metamaterial optical response with semiconductor quantum dots," *Opt. Express* **17**(10), 8548–8551 (2009), <http://www.opticsinfobase.org/oe/abstract.cfm?URI=oe-17-10-8548>.
10. N. Meinzer, M. Ruther, S. Linden, C. M. Soukoulis, G. Khitrova, J. Hendrickson, J. D. Oltzky, H. M. Gibbs, and M. Wegener, "Arrays of Ag split-ring resonators coupled to InGaAs single-quantum-well gain," *Opt. Express* **18**(23), 24140–24151 (2010), <http://www.opticsinfobase.org/oe/abstract.cfm?URI=oe-18-23-24140>.
11. D. J. Bergman and M. I. Stockman, "Surface plasmon amplification by stimulated emission of radiation: quantum generation of coherent surface plasmons in nanosystems," *Phys. Rev. Lett.* **90**(2), 027402 (2003).
12. M. A. Noginov, G. Zhu, A. M. Belgrave, R. Bakker, V. M. Shalaev, E. E. Narimanov, S. Stout, E. Herz, T. Suteewong, and U. Wiesner, "Demonstration of a spaser-based nanolaser," *Nature* **460**(7259), 1110–1112 (2009).
13. M. I. Stockman, "Spasers explained," *Nat. Photonics* **2**(6), 327–329 (2008).
14. M. I. Stockman, "Ultrafast nanoplasmonics under coherent control," *New J. Phys.* **10**(2), 025031 (2008).
15. M. I. Stockman, "The spaser as a nanoscale quantum generator and ultrafast amplifier," *J. Opt.* **12**(2), 024004 (2010).
16. I. R. Gabitov, B. Kennedy, and A. I. Maimistov, "Coherent amplification of optical pulses in metamaterials," *IEEE J. Sel. Top. Quantum Electron.* **16**(2), 401–409 (2010).
17. V. V. Klimov, "Nanoplasmonics," *Phys. Usp.* **51**(8), 839–844 (2008).
18. L.-M. Si, T. Jiang, K. Chang, T.-C. Chen, X. Lv, L. Ran, and H. Xin, "Active microwave metamaterials incorporating ideal gain devices," *Materials* **4**(1), 73–83 (2011).
19. A. D. Boardman, Y. G. Rapoport, N. King, and V. N. Malnev, "Creating stable gain in active metamaterials," *J. Opt. Soc. Am. B* **24**(10), A53–A61 (2007).

20. Y.-Y. Yu, D.-Z. Lin, L.-S. Huang, and C.-K. Lee, "Effect of subwavelength annular aperture diameter on the nondiffracting region of generated Bessel beams," *Opt. Express* **17**(4), 2707–2713 (2009), <http://www.opticsinfobase.org/oe/abstract.cfm?URI=oe-17-4-2707>.
21. A. Govyadinov, V. Podolskiy, and M. A. Noginov, "Active metamaterials: Sign of refractive index and gain-assisted dispersion management," *Appl. Phys. Lett.* **91**(19), 191103 (2007).
22. A. Muller, Q. Q. Wang, P. Bianucci, C. K. Shih, and Q. K. Xue, "Determination of anisotropic dipole moments in self-assembled quantum dots using Rabi oscillations," *Appl. Phys. Lett.* **84**(6), 981–983 (2004).
23. L. Novotny and B. Hecht, *Principles of Nano-Optics* (Cambridge University Press, 2006).
24. R. H. Pantell and H. E. Puthoff, *Fundamentals of Quantum Electronics* (Wiley, 1969).
25. P. W. Milonni, *The Quantum Vacuum: An Introduction to Quantum Electrodynamics* (Academic Press, 1994).
26. F. Wang and Y. R. Shen, "General properties of local plasmons in metal nanostructures," *Phys. Rev. Lett.* **97**(20), 206806 (2006).
27. M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, 1997).
28. I. E. Protsenko, A. V. Uskov, O. A. Zaimidoroga, V. N. Samoilov, and E. P. O'Reilly, "Dipole nanolaser," *Phys. Rev. A* **71**(6), 063812 (2005).
29. K. Li, X. Li, M. I. Stockman, and D. J. Bergman, "Surface plasmon amplification by stimulated emission in nanolenses," *Phys. Rev. B* **71**(11), 115409 (2005).
30. A. S. Rosenthal and T. Ghannam, "Dipole nanolasers: A study of their quantum properties," *Phys. Rev. A* **79**(4), 043824 (2009).
31. A. N. Lagarkov, A. K. Sarychev, V. N. Kissel, and G. Tartakovsky, "Superresolution and enhancement in metamaterials," *Phys. Usp.* **52**(9), 959–967 (2009).
32. A. Pikovsky, M. Rosenblum, and J. Kurths, *Synchronization. A Universal Concept in Nonlinear Sciences* (Cambridge University Press, 2001).

## 1. Introduction

Joule (ohmic) losses in metallic nano-inclusions embedded in a host metamaterial medium are the main obstacle to realizing a practical "perfect" lens with subwavelength resolution as well as to other applications of metamaterials [1]. Losses can be compensated for, however, in amplifying media. The authors of [2,3] proposed making a stackable lens with alternating metamaterial and gain layers. This can be accomplished by directly introducing gain inclusions such as molecules or quantum dots (QDs) into the matrix [4–6]. The latter scheme has recently been realized experimentally [7–10] demonstrating that it is possible to compensate for losses.

The combination of a gain medium and metallic nanoparticles (NPs) results in the emergence of a spaser (Surface Plasmon Amplification by Stimulated Emission of Radiation) first suggested by Bergman and Stockman [11] and realized experimentally in [12]. Spasers, as well as nanoplasmonics overall, is a new fast growing field with tremendous promise. Schematically, the spaser is a system of inversely excited two-level QDs surrounding metal nanoparticles [11,13]. Its principles of operation are analogous to those for a laser with the role of photons played by surface plasmons (SPs) localized at a NP that serves as the resonator [11,14,15]. In other words, in a spaser, near-fields of the NP are generated and amplified. The amplification of the SPs occurs due to non-radiative energy transfer from QDs. This process takes place due to the dipole-dipole (or any other near-field [16]) interaction between the QD and the plasmon NP. The efficiency of such a mechanism depends on the probability of the non-radiative excitation of the surface plasmon, which is  $(kr)^{-3}$  greater than the radiation of the photon [17], where  $r$  is the distance between the centers of the QD and the NP and  $k$  is the photon wavenumber. Stimulated radiation from the QD into the plasmon mode results in spasing. The excitation of the plasmon mode is carried out via the excitation of the QD.

In theoretical studies of a metamaterial with compensated losses, it is assumed that electromagnetic wave propagation can be described by the Maxwell equations with real valued effective electric permittivity and magnetic permeability. In its turn, the use of effective permittivities and refraction coefficients assumes that the dipole moment of a NP oscillates with the frequency of the external field and the amplitude of this oscillation is determined by the external field. In the absence of the external field, there should be no oscillations of a dipole moment. The classical linear description treats the gain medium as a system with a negative imaginary part of the permittivity [18–21].

However, when spasers are used as gain inclusions, their dipole moments are excited not only by the external field but also by the radiation produced by QDs, which actually tends to compensate for loss. Unlike the classical linear description [18–21], the semi-classical analysis shows that the spaser in the presence of pumping is a self-oscillating system. The dipole moment of the spaser's NP oscillates autonomically even in the absence of the external field. The autonomic frequency of this self-oscillation is determined by the plasmon frequency, transition frequency of the gain inclusion and characteristic times of relaxation in NP and excitation of gain inclusions [15]. Therefore, in order to develop a correct description of metamaterials with spasers, it is necessary to study the interaction of the spaser with the external field in details.

In this paper, we study analytically and numerically the operation of a spaser driven by an external optical wave. We demonstrate that the pumping drastically changes the spaser's behavior in the optical field in comparison with the behavior of a passive QD-NP pair. In particular, when the frequency of the external field is detuned from the autonomous frequency of the spaser, even infinitesimally weak field drives the spaser to stochastic oscillations. The spaser can be synchronized to the external field only when the field amplitude exceeds a threshold value.

## 2. Equations of motion of the spaser

The simplest model of the spaser consists of a two-level QD of size  $r_{TLS}$  which is positioned at a distance  $r$  from a metallic NP of size  $r_{NP}$  [11]. The whole system is immersed into a solid dielectric or semiconductor matrix with dielectric permittivity  $\varepsilon_M$ . The dipole moment of a typical QD of size  $r_{TLS} \approx 10$  nm is  $\mu_{TLS}^{class} \sim 20$  Debye [22]. For a NP with Ag-core and SiO<sub>2</sub>-shell with radius  $r_{NP} \approx r \approx 10$  nm, the classical dipole moment near the frequency of the plasmon resonance,  $\omega$ , can be estimated to be  $\mu_{NP}^{class} \sim 200$  Debye. The energy of the NP-QD quasistatic dipole-dipole interaction is estimated to be  $V = \hbar\Omega_R \sim \mu_{NP}^{class} \mu_{TLS}^{class} / r^3$ , which gives  $\Omega_R \approx 5 \cdot 10^{12} \text{ s}^{-1} \sim 10^{-2} \omega$ . Thus, the Rabi frequency (the coupling constant) is two orders of magnitude smaller than the generation frequency. This allows for the use of the slowly varying envelope approximation in the investigation of the dynamics of the spaser.

The transition processes of the NP and two-level QD interacting in a spaser can be described by the model Hamiltonian

$$\hat{H} = \hat{H}_{sp} + \hat{H}_{TLS} + \hat{V} + \hat{\Gamma}, \quad (1)$$

where

$$\hat{H}_{sp} = \hbar\omega_{sp} \hat{a}^\dagger(t) \hat{a}(t), \quad (2)$$

$$\hat{H}_{TLS} = \hbar\omega_{TLS} \hat{\sigma}^\dagger(t) \hat{\sigma}(t) \quad (3)$$

describe the non-interacting NP and QD, respectively [11,23,24], the operator  $\hat{V} = -\hat{\mu}_{TLS} \cdot \hat{\mathbf{E}}_{NP}$  determines their interaction. To take into account the interaction of the spaser, which is an open system, with surroundings (pumping, phonons in metal, etc.) following [24] we introduce the non-Hermitian operator  $\hat{\Gamma}$  that describes relaxation and pumping processes. Here  $\hat{a}(t)$  is the annihilation operator of the dipole SP,  $\hat{\mu}_{TLS} = \mu_{TLS} (\hat{\sigma}(t) + \hat{\sigma}^\dagger(t))$  is the operator for the dipole moment of the QD,  $\hat{\sigma} = |g\rangle\langle e|$  is the transition operator between ground  $|g\rangle$  and excited  $|e\rangle$  states of the QD,  $\mu_{TLS} = \langle e|e\mathbf{r}|g\rangle$  is the QD dipole moment

matrix element. Quantization of the plasmon field without taking into account losses is carried out in the standard way [25]. Thus, we obtain [11,15]

$$\hat{\mathbf{E}}_{NP} = \sum_q \sqrt{\frac{\hbar \omega_q}{2W_q}} \left[ \hat{a}_q \mathbf{E}_q(\mathbf{r}) \exp(-i\omega_q t) + \hat{a}_q^* \mathbf{E}_q^*(\mathbf{r}) \exp(i\omega_q t) \right], \quad (4)$$

where

$$W_q = \frac{1}{4\pi} \int \frac{\partial(\text{Re } \varepsilon \omega^2)}{\partial \omega^2} \bigg|_{\omega_q} E_q E_q^* dV = \frac{1}{8\pi} \int \left( 2\text{Re } \varepsilon + \omega \frac{\partial \text{Re } \varepsilon}{\partial \omega} \right) \bigg|_{\omega_{SP}} E_1 E_1^* dV \quad (5)$$

is the normalization factor and  $\mathbf{E}_q(\mathbf{r}) = -\nabla \varphi_q$  is the  $q$ -th eigenmode determined by the geometry of the problem [11]. In the dipole approximation, in Eq. (4) we retain only one dipole mode ( $q=1$ ) with the frequency  $\omega_{SP} = \omega_1$ . For a spherical NP this dipole mode is uniform inside the particle and has the form of the field of the dipole with a unitary dipole moment,  $\boldsymbol{\mu}_1$ , so that  $\mathbf{E}_1 = -\boldsymbol{\mu}_1 r^{-3} + 3(\boldsymbol{\mu}_1 \cdot \mathbf{r})\mathbf{r}r^{-5}$  and  $\mathbf{E}_1 = -\boldsymbol{\mu}_1 r_{NP}^{-3}$  outside and inside of the NP, respectively. Note, that even though  $\boldsymbol{\mu}_1$  is a unit vector, it is a dimensional quantity, therefore, below we will show  $|\boldsymbol{\mu}_1|$  explicitly.

In the absence of the external field, for the NP near field we have  $(8\pi)^{-1} \int \text{Re } \varepsilon|_{\omega_{SP}} E_1 E_1^* dV = 0$  [26]. The near field is not equal to zero at the resonance frequency only. However, in this case, the field vanishes at the infinity and  $\int \text{Re } \varepsilon|_{\omega_{SP}} E_1 E_1^* dV = \int \text{Re } \varepsilon|_{\omega_{SP}} \nabla \varphi^* \cdot \nabla \varphi dV = -\int \varphi^* \nabla \cdot (\text{Re } \varepsilon|_{\omega_{SP}} \nabla \varphi) dV$ . The latter integral is equal to zero because  $\nabla^2 \varphi = 0$  both inside and outside the NP. Furthermore,  $\partial \varepsilon / \partial \omega = 0$  outside of the particle. This leads to the following result

$$W_1 = \frac{1}{8\pi} \int_{\text{volume of NP}} \omega \frac{\partial \text{Re } \varepsilon}{\partial \omega} \bigg|_{\omega_{SP}} E_1 E_1^* dV = \frac{|\boldsymbol{\mu}_1|^2}{6r_{NP}^3} \omega \frac{\partial \text{Re } \varepsilon}{\partial \omega} \bigg|_{\omega_{SP}}, \quad (6)$$

which gives  $\hat{\mathbf{E}}_{NP} = \sqrt{3\hbar r_{NP}^3 / (|\boldsymbol{\mu}_1|^2 \partial \text{Re } \varepsilon / \partial \omega)} \mathbf{E}_1(\mathbf{r}) (\hat{a} + \hat{a}^\dagger)$ . Thus, the operator for the dipole moments of the NP is  $\hat{\boldsymbol{\mu}}_{NP} = \boldsymbol{\mu}_{NP} (\hat{a} + \hat{a}^\dagger)$ , where  $\boldsymbol{\mu}_{NP} = \sqrt{3\hbar r_{NP}^3 / (\partial \text{Re } \varepsilon_M / \partial \omega)} \boldsymbol{\mu}_1 / |\boldsymbol{\mu}_1|$

Assuming that the frequencies of the QD transition and the frequency of the dipole SP are close,  $\omega_{SP} \approx \omega_{TLS}$ , we will look for the time dependencies of  $\hat{a}(t)$  and  $\hat{\sigma}(t)$  in the form  $\hat{a}(t) = \hat{a}(t) \exp(-i\omega_a t)$  and  $\hat{\sigma}(t) = \hat{\sigma}(t) \exp(-i\omega_a t)$ , where  $\hat{a}(t)$  and  $\hat{\sigma}(t)$  are the slowly changing amplitudes and  $\omega_a$  is the autonomous frequency of the spaser which we seek. In the rotating wave approximation [24] we can neglect rapidly oscillating terms,  $\propto \exp(\pm 2i\omega_a t)$ , and obtain the interaction operator  $\hat{V}$  in the form of the Jaynes–Cummings Hamiltonian [27]

$$\hat{V} = \hbar \Omega_R (\hat{a}^\dagger \hat{\sigma} + \hat{\sigma}^\dagger \hat{a}), \quad (7)$$

where  $\Omega_R = -\sqrt{3r_{MP}^3 / (\partial \text{Re } \varepsilon / \partial \omega)} \mathbf{E}_1(\mathbf{r}) \cdot \boldsymbol{\mu}_{TLS} / |\boldsymbol{\mu}_1|$  is the coupling constant (see also [11,28]).

The commutation relations for operators  $\hat{a}(t)$  and  $\hat{\sigma}(t)$  are standard:  $[\hat{a}, \hat{a}^\dagger] = \hat{1}$  and  $[\hat{\sigma}^\dagger, \hat{\sigma}] = \hat{D}$ , where the operator  $\hat{D}$  describes the inversion of the occupancies of the ground and excited states of the QD,  $\hat{D}(t) = \hat{n}_e(t) - \hat{n}_g(t)$ ,  $\hat{n}_e = |e\rangle\langle e|$  and  $\hat{n}_g = |g\rangle\langle g|$  are the occupancy operators for the ground and excited states,  $\hat{n}_g + \hat{n}_e = \hat{1}$ . Using the Hamiltonian, Eqs. (1)-(3), and the commutation relations for operators  $\hat{a}(t)$  and  $\hat{\sigma}(t)$  we obtain the Heisenberg equations of motion for the operators  $\hat{a}(t)$ ,  $\hat{\sigma}(t)$ , and  $\hat{D}(t)$  [28,29]

$$\dot{\hat{D}} = 2i\Omega_R(\hat{a}^\dagger \hat{\sigma} - \hat{\sigma}^\dagger \hat{a}) - \frac{\hat{D} - \hat{D}_0}{\tau_D}, \quad (8)$$

$$\dot{\hat{\sigma}} = (i\delta_{TLS} - \frac{1}{\tau_\sigma})\hat{\sigma} + i\Omega_R \hat{a} \hat{D}, \quad (9)$$

$$\dot{\hat{a}} = (i\delta_{SP} - \frac{1}{\tau_a})\hat{a} - i\Omega_R \hat{\sigma}, \quad (10)$$

where  $\delta_{TLS} = \omega_a - \omega_{TLS}$  and  $\delta_{SP} = \omega_a - \omega_{SP}$  are the frequency differences. The Markovian interaction with the reservoir is described by the Liouvillian (non-Hermitian operator) term  $\hat{\Gamma}$  determining the decay rates with relaxation times  $\propto \tau_D^{-1}$ ,  $\tau_\sigma^{-1}$ , and  $\tau_a^{-1}$ , which account for the relaxation processes for the SP annihilation operator, the QD polarization and the occupancy operators, respectively [27]. The operator for the occupancy inversion  $\hat{D}_0$  describes the pumping [24,27]. We now neglect quantum fluctuations and correlations and consider  $\hat{a}(t)$ ,  $\hat{\sigma}(t)$  and  $\hat{D}(t)$  as complex quantities (c-numbers), so that we can use complex conjugation instead of Hermitian conjugation [11,28,30,31]. The difference between the occupancies of the upper and lower levels  $D(t)$  must be a real valued quantity because the respective operator is Hermitian. The quantities  $\sigma(t)$  and  $a(t)$  are the complex amplitudes of the dipole oscillations of the QD and SP, respectively.

For  $D_0 < D_{th}$  the system of Eqs. (8)-(10) has a trivial stationary solution  $a = \sigma = 0$ ,  $D = D_0$  only, while for  $D_0 > D_{th}$  there are two stationary solutions. The trivial unstable solution corresponds to the absence of SPs, while the stable one corresponds to laser generation:

$$a = \frac{e^{i\varphi}}{2} \sqrt{\frac{(D_0 - D_{th})\tau_a}{\tau_D}}, \quad \sigma = \frac{e^{i\psi}}{2} \sqrt{\frac{(D_0 - D_{th})(\delta_{SP}^2 + 1/\tau_a^2)\tau_a}{\Omega_R^2 \tau_D}}, \quad (11)$$

$$D = D_{th}, \quad \omega = \frac{\omega_{SP}\tau_a + \omega_{TLS}\tau_\sigma}{\tau_a + \tau_\sigma},$$

where  $\exp(i(\psi - \varphi)) = (\delta_{SP} + i/\tau_a)/(\delta_{SP}^2 + 1/\tau_a^2)^{1/2}$ . The unstable solution appears only when the pumping level reaches the threshold value  $D_{th} = (1 + \delta_{SP}^2 \tau_a^2)/(\Omega_R^2 \tau_a \tau_\sigma)$  (see Fig. 1). The stationary values of  $a$ ,  $\sigma$ , and  $D$  shown in Fig. 1.

### 3. The spaser in the external electromagnetic field

Let us consider the dynamics of the NP and QD in the field of the external optical wave,  $E(t) = E \cos \omega_f t$ . Assuming that the external field is classical and taking into account the dipole interaction only, we can write the Hamiltonian in the form

$$\begin{aligned} \hat{H}_{ef} = & \hat{H} + \hbar\Omega_1 (\hat{a}^\dagger + \hat{a}) (\exp(i\omega_f t) + \exp(-i\omega_f t)) \\ & + \hbar\Omega_2 (\hat{\sigma}^\dagger + \hat{\sigma}) (\exp(i\omega_f t) + \exp(-i\omega_f t)), \end{aligned} \quad (12)$$

where  $\hat{H}$  is defined by Eq. (2),  $\Omega_1 = -\mu_{NP} E / \hbar$ , and  $\Omega_2 = -\mu_{TLS} E / \hbar$ . As before, we use the Heisenberg equations for operators  $\hat{a}$ ,  $\hat{\sigma}$ , and  $\hat{D}$  to obtain equations of motion for “slowly varying” amplitudes,

$$\dot{\hat{D}} = 2i\Omega_R (\hat{a}^\dagger \hat{\sigma} - \hat{\sigma}^\dagger \hat{a}) + 2i\Omega_2 (\hat{\sigma} - \hat{\sigma}^\dagger) - \frac{\hat{D} - \hat{D}_0}{\tau_D}, \quad (13)$$

$$\dot{\hat{\sigma}} = (i\Delta_{TLS} - \frac{1}{\tau_\sigma}) \hat{\sigma} + i\Omega_R \hat{a} \hat{D} + i\Omega_2 \hat{D}, \quad (14)$$

$$\dot{\hat{a}} = (i\Delta_{SP} - \frac{1}{\tau_a}) \hat{a} - i\Omega_R \hat{\sigma} - i\Omega_1. \quad (15)$$

Below we assume for simplicity that  $\omega_{TLS} = \omega_{SP}$ . Hence  $\Delta_{SP} = \omega_f - \omega_{SP} = \Delta_{TLS} = \omega_f - \omega_{TLS} = \Delta$ . Note that without the QD, the polarization of the NP corresponding to the stationary solution of the system of Eqs. (13)-(15),  $\dot{\hat{a}} = \dot{\hat{\sigma}} = 0$ , has the form

$$\alpha r_{NP}^{-3} = \frac{3}{2(\Delta_{SP} + i/\tau_a)} \left( \frac{\partial \varepsilon_{NP}(\omega)}{\partial \omega} \right)^{-1}, \quad (16)$$

where  $\varepsilon_{NP}(\omega)$  is the permittivity of the metal NP and  $\alpha$  is its polarizability. Generally,  $\varepsilon_{NP}(\omega)$  is an effective permittivity of the NP in the matrix. However, for large enough NPs it can be considered as the bulk permittivity of the NP's metal. In the slowly varying amplitude approximation, i.e. for a small detuning  $\Delta_{SP} \ll 1$ , this expression coincides with the classical one  $\alpha^{class} r_{NP}^{-3} = [\varepsilon_{NP}(\omega) - \varepsilon_M] / [\varepsilon_{NP}(\omega) + 2\varepsilon_M]$  (see also [30]).

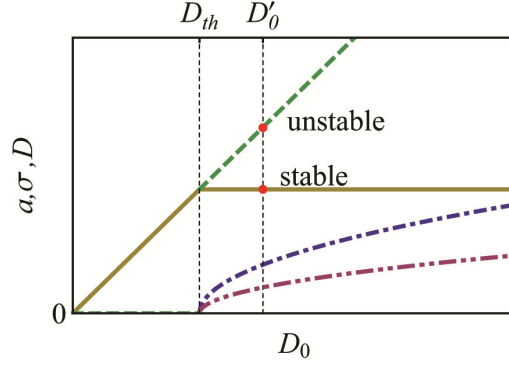


Fig. 1. Stationary amplitudes  $a$  and  $\sigma$  are shown by dash-double dotted and dash-dotted lines, respectively. The stable solution for  $D$  is shown by the solid line. The unstable solution appearing for  $D > D_{th}$  is shown by the dashed line. For  $D_0 = D'_0$  (also shown in Fig. 2) the stable and unstable solutions of  $D$  are marked by red dots.

The quantities defined by Eqs. (13)-(15) differ from those in Eqs. (9)-(11). Without the external field the frequency  $\omega_a$  in the substitutions  $\hat{\hat{a}}(t) = \hat{a}(t)\exp(-i\omega_a t)$  and  $\hat{\hat{\sigma}}(t) = \hat{\sigma}(t)\exp(-i\omega_a t)$  is found from the solution of stationary Eqs. (8)-(10). When the external field is present, we use the representations  $\hat{\hat{a}}(t) = \hat{a}(t)\exp(-i\omega_f t)$  and  $\hat{\hat{\sigma}}(t) = \hat{\sigma}(t)\exp(-i\omega_f t)$ . We look for a stationary solution of the system of Eqs. (13)-(15). The stationary values of  $D$  satisfy a cubic equation and are shown in Fig. 2 as a function of the amplitude of the external field  $E$ . For  $\Delta = 0$  (Fig. 2a) in the absence of the external field  $E = 0$  and  $D_0 \geq D_{th}$  two points, stable and unstable, correspond to the points marked by dots in Fig. 1.

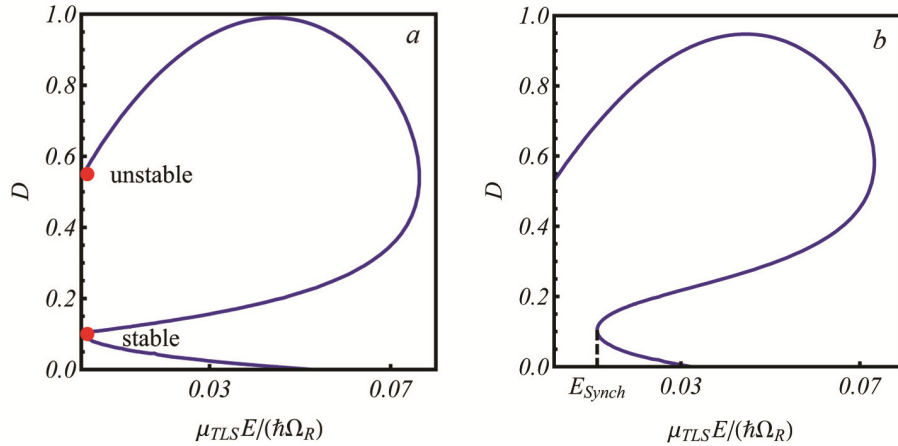


Fig. 2. The stationary values of  $D$  as a function of the amplitude of the external field for (a) zero ( $\Delta = 0$ ) and (b) non-zero ( $\Delta = 10^{11} \text{ s}^{-1}$ ) detuning. For both graphs  $\tau_a = 10^{-14} \text{ s}$ ,  $\tau_\sigma = 10^{-11} \text{ s}$ ,  $\tau_D = 0.5 \cdot 10^{-14} \text{ s}$ ,  $\Omega_R = 10^{13} \text{ s}^{-1}$ ,  $D_0 = D'_0 = 0.55$ .

#### 4. Spaser synchronization

Only the lower branch of the synchronization region corresponds to a stable solution. The stationary solutions at zero external field and with no detuning ( $\delta_{TLS} = \delta_{sp} = \Delta = 0$ ) are shown

in Fig. 1. When detuning is present (we still assume that  $\delta_{SP} = \delta_{TLS} = 0$  but  $\Delta \neq 0$ ), the stable stationary solution exists for  $E > E_{synch}(\Delta)$  only (Fig. 2b). This result is confirmed by our numerical simulation shown in Fig. 3. Such behavior of a self-oscillating system is referred to as synchronization by an external periodic influence [32]. The region in which the synchronization takes place is known as the Arnold tongue. To obtain an analytical estimate of the Arnold tongue boundary, we find the solution of Eqs. (13)-(15) in the first approximation with respect to the field. For the case  $\delta_{SP} = 0$ , using the substitution  $a = |a|\exp(i\varphi)$  and  $\sigma = |\sigma|\exp(i\psi)$  in Eq. (15) and equating its imaginary parts, we obtain

$$\dot{\varphi} = \Delta - \Omega_R \frac{|\sigma|}{|a|} \cos(\psi - \varphi) - \frac{\Omega}{|a|} \cos \varphi. \quad (17)$$

Since  $E(t) = E \cos \omega_f t$ ,  $\varphi$  represents the phase shift between the external field and spaser oscillations. For  $|a|$ ,  $|\sigma|$ , and  $\cos(\psi - \varphi)$  we can use their values obtained from Eq. (11) in the zeroth order with respect to the field approximation. As the result, we have

$$\dot{\varphi} = \Delta - \xi \cos \varphi, \quad (18)$$

where  $\xi = \Omega_1 / |a|$  (in this approximation  $\cos(\psi - \varphi) = 0$ ). If one introduces a function  $U(\varphi) = \Delta[-\varphi + (\xi \sin \varphi) / \Delta]$ , Eq. (18) takes the form of Adler's equation [32]:

$$\dot{\varphi} = -\frac{\partial U(\varphi)}{\partial \varphi}. \quad (19)$$

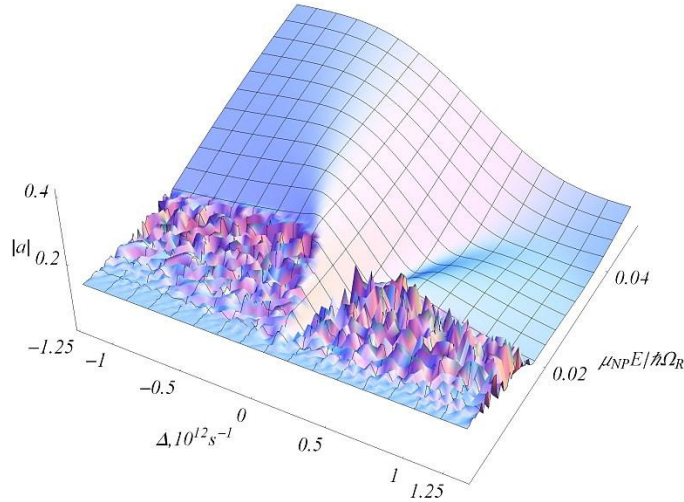


Fig. 3. The dependence of the plasmon dipole moment on the amplitude of the external field and the frequency detuning  $\Delta$ . The speckle structure at low values of  $E$  corresponds to the chaotic behavior of the dipole moment.

The phase dynamics can be viewed as the motion of a particle sliding along the potential profile  $U(\varphi)$  (Fig. 4) in a viscous liquid. For a small field and/or large detuning,  $|\xi / \Delta| < 1$ , the phase difference of the system and the external field increases monotonically. For  $|\xi / \Delta| > 1$ , the “particle” should be trapped in one of the minima of the potential function. In this regime the oscillation of the system is synchronized and the phase difference is time

independent. In the approximation considered, the width of the synchronization region is proportional to the field amplitude. Within the Arnold tongue, if the energy of the interaction of the NP with the external field is smaller than the QD-NP interaction, the amplitude of the auto-oscillations depends weakly upon the amplitude of the external field, which in this case plays the role of a synchronizer.

When  $\Delta \rightarrow 0$  the threshold field value  $E_{synch}$  is determined by the equation  $(\mu_{NP} E_{synch} / \hbar)^2 = (D_0 - D_{th}) \Delta^2 (\tau_a / 4\tau_D)$ . For large values of the detuning,  $E_{synch}$  becomes independent of  $\Delta$  (see Fig. 3). Taking into account that  $\tau_a \sim 10^{-14} s^{-1}$  and  $\tau_\sigma \sim 10^{-11} s^{-1}$  for plasmonic NPs, one can estimate the asymptotic value of the Arnold tongue boundary,  $E_{synch}^*$ . Our numerical calculations show that  $E_{synch}(\Delta)$  tends to a plateau for  $\Delta \sim 2 \cdot 10^{11} s^{-1}$ , which gives  $E_{synch}^* \sim 3 \cdot 10^3 V/m$ . This agrees with calculations of [3,10] in which wave propagation in the system of spasers was considered. In [3,10] the amplitude of the incident wave was a few orders of magnitude larger than  $E_{synch}^*$ . Such a field is comparable with the near field inside the spaser. In this case, spaser becomes synchronized with the external field for any value of detuning; it ceases to be an autonomous system and responds linearly to the external field as can be seen in Fig. 3. For such strong fields, losses cannot be compensated exactly. Moreover, our numerical calculations show that for a very strong field, the population inversion of the QD decreases, the excitation of SPs by QDs is inhibited, and the spaser becomes just a lossy NP. Thus, when detuning is substantial, spasers cannot be used as active inclusions for the perfect lens because both loss and amplification destroy the perfect image, so that the exact compensation of loss is required.

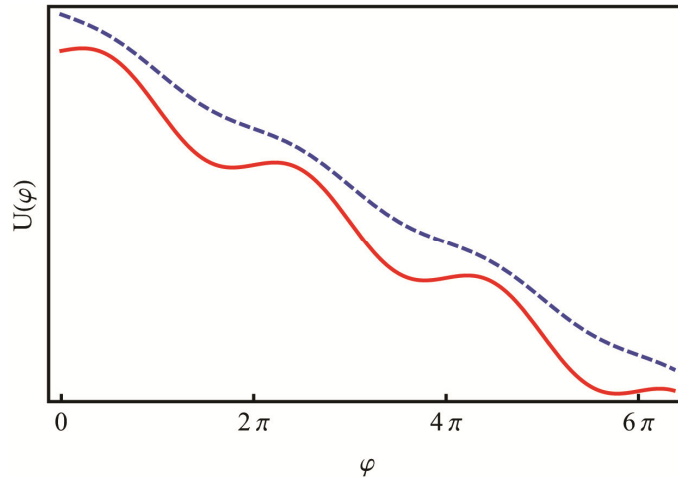


Fig. 4. The potential  $U(\varphi)$  for  $|\xi / \Delta| < 1$  (dashed blue line) and  $|\xi / \Delta| > 1$  (solid red line).

For applications it is important to know not only a response of a single spaser to an external electromagnetic wave but also the behavior of a system of interacting spasers randomly or regularly distributed in the dielectric matrix. The interaction between spasers may lead to a cooperative behavior of spasers. This problem is a subject of our future research.

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