

Switching waves and dissipative structures in a chain of spasers

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Abstract. We have considered the physical mechanism of optical bistability in a spaser in the field of an external optical wave. We have studied the effect of this phenomenon on the dynamics of a 1D chain of coupled spasers. It is shown that such a chain demonstrates the behaviour typical of open nonlinear systems. In particular, for high Joule losses in a spaser, a nonlinear switching autowave propagates in the chain, thereby evolving the spasers' state from a low population inversion state into a high population inversion state or vice versa. The control parameter that determines the type of switching is the amplitude of the external optical wave. For low Joule losses there emerge quasi-periodic dissipative structures whose formation dynamics is of 'self-assembly' nature.

Keywords: *nanoplasmonics, spasers, bistability, switching waves, dissipative structures, self-assembly.*

5. Introduction

Recently, a new field of optics, quantum plasmonics, which combines the advantages of plasmonics and quantum electronics [1–25] has experienced an explosive growth. Plasmonics operates with surface waves, whose wavelengths are much smaller than an optical wavelength in vacuum. This endows plasmonics with many features of near-field optics and makes it popular with modern nanotechnologies. We may mention here SERS, spasers, nanoscale light sources [26–30], and various devices based on metamaterials [17, 31, 32]: nanoscale energy concentrators and transmission lines, superlenses with a resolution that exceeds the diffraction limit, devices masking nanoobjects (clocking), hyperlenses [33–40], etc.

The smallness of scales, on which phenomena specific to plasmonics are developing, leads to the need to take quantum effects into account. In this connection there appears a new discipline, i.e., quantum plasmonics, which considers quantum effects under conditions of plasmon resonance.

The objects of quantum plasmonics arousing considerable interest recently are nanolasers, which include a dipole nano-

laser [8, 10], spaser [11, 41], a nanolaser based on a magnetic plasmon resonance mode [42, 43]. The experimental realisation of a spaser has been recently reported in [44]. The principle of spaser operation is similar to that of a laser, i.e., the gain is provided by the inverse population in combination with the feedback ensured by stimulated emission of a quantum dot. In a spaser, the role of photons is played by surface plasmons of nanoparticles. Their localisation on a nanoparticle [11, 41, 45] creates prerequisites for the implementation of feedback. In other words, the spaser operation is accompanied by generation and amplification of near fields of a nanoparticle (spasing). Amplification of surface plasmons is due to nonradiative energy transfer from the quantum dot. At the heart of the process is the dipole–dipole (or any other near-field [46]) interaction of a quantum dot and plasmonic nanoparticle. This mechanism can be considered as the principal one, because the probability of nonradiative plasmon excitation is $(kr_0)^{-3}$ times higher than radiative emission of a photon [15] (r_0 is the distance between the centres of the nanoparticle and quantum dot, $k = 2\pi/\lambda$, λ is the wavelength in vacuum). Thus, the efficiency of energy transfer from the quantum dot to the surface plasmon of the nanoparticle is caused by a small distance r_0 , despite the fact that the Q factor of the plasmon resonance is quite low*. Due to high efficiency of this process, an external optical wave propagating in a composite of spasers interacts not separately with quantum dots and nanoparticles but with whole spasers.

Above the lasing threshold, a spaser (like a laser) is a self-oscillating system. The spaser's behaviour in this regime (synchronisation by an external field, compensation for loss, non-equilibrium response, etc.) is considered in detail in [20–22, 24, 25]. Below we study the behaviour of spasers under low external pump when spasing does not emerge yet. Protsenko et al. [7] showed that under the action of an external field a spaser operating below the threshold may exhibit bistable behaviour. That is, for each value of the field there are two stable steady states of a spaser with different population inversions of the quantum dot. Kaplan and Volkov [49, 50] predicted the formation of structures in a chain of nonlinear two-level atoms resonantly interacting with the external field. In this paper, we study the dynamics of a system, which has, along with the resonance line of the quantum dot transition, a plasmon resonance of a nanoparticle, i.e., we consider a chain of bistable spasers. We also show that in such a chain, along with the formation of dissipative structures, switching waves can propagate.

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*Note that we neglect below the processes of photon emission; in this case, the Purcell effect [47, 48] does not play any noticeable role and can be ignored.

6. Bistability of a single spaser in an external field

The dynamics of a nanoparticle and quantum dot in an external optical wave field $E(t) = E \cos(\nu t)$, which can be considered classical, allows one to restrict consideration to dipole interaction only and is described by a Hamiltonian [20, 21, 25]:

$$\begin{aligned} \hat{H} = & \hat{H}_0 + \hbar\Omega_R(\hat{a}^\dagger \hat{\sigma} + \hat{\sigma}^\dagger \hat{a}) + \hbar\Omega_1(\hat{a}^\dagger + \hat{a})[\exp(i\nu t) + \exp(-i\nu t)] \\ & + \hbar\Omega_2(\hat{\sigma}^\dagger + \hat{\sigma})[\exp(i\nu t) + \exp(-i\nu t)], \end{aligned} \quad (1)$$

where $\hat{H}_0 = \hbar\omega_{\text{NP}}\hat{a}^\dagger\hat{a} + \hbar\omega_{\text{TLS}}\hat{\sigma}^\dagger\hat{\sigma}$ is a Hamiltonian of a non-interacting nanoparticle and a quantum dot, and $\Omega_1 = -\mu_{\text{NP}}E/\hbar$ and $\Omega_2 = -\mu_{\text{TLS}}E/\hbar$ are the coupling constants with the external field; $\mu_{\text{TLS}} = \langle e | \hat{d}_{\text{TLS}} | g \rangle$ is the transition dipole moment of a quantum dot; $\mu_{\text{NP}}^2 = 3\hbar r_{\text{NP}}^3/(\partial \text{Re} \epsilon_{\text{NP}}/\partial \omega)$ [20]; r_{NP} and ϵ_{NP} are the radius and the permittivity of a nanoparticle.

Using the Heisenberg equation, for the ‘slow’ amplitudes \hat{a} , $\hat{\sigma}$ and \hat{D} we obtain the equations:

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

$$\dot{\hat{\sigma}} = \left(i\delta - \frac{1}{\tau_\sigma}\right)\hat{\sigma} + i\Omega_R\hat{a}\hat{D} + i\Omega_2\hat{D}, \quad (3)$$

$$\dot{\hat{a}} = \left(i\Delta - \frac{1}{\tau_a}\right)\hat{a} - i\Omega_R\hat{\sigma} - i\Omega_1, \quad (4)$$

where τ_D and τ_σ are the longitudinal and transverse relaxation times of a quantum dot; τ_a is the lifetime of a nanoparticle plasmon; $\Omega_R = [\mu_{\text{NP}}\mu_{\text{TLS}} - 3(\mu_{\text{NP}}\mathbf{e}_r)(\mu_{\text{NP}}\mathbf{e}_r)]/\hbar r_0^3$ is the Rabi frequency; $\mathbf{e}_r = \mathbf{r}/r$ is a unit vector of the direction of a quantum dot to a nanoparticle; $\delta = \nu - \omega_{\text{TLS}}$; $\Delta = \nu - \omega_{\text{SP}}$ is the frequency detuning; ω_{TLS} is the transition frequency of a quantum dot; ω_{SP} is the plasmon resonance frequency; ν is the frequency of the external field. We are interested in conditions under which there appears bistability of a spaser interacting with the external field.

The bistability phenomenon of a single spaser was first considered in [7]. To understand the physical mechanism of this phenomenon, we consider the steady-state solution of this phenomenon, we consider the steady-state solution of system (2)–(4). We set the time derivatives equal to zero and pass from the operators to c -numbers. As a result, we obtain the system of equations:

$$2i\Omega_R(a^* \sigma - \sigma^* a) + 2i\Omega_2(\sigma - \sigma^*) - \frac{D - D_0}{\tau_D} = 0, \quad (5)$$

$$\left(i\delta - \frac{1}{\tau_\sigma}\right)\sigma + i\Omega_R a D + i\Omega_2 D = 0, \quad (6)$$

$$\left(i\Delta - \frac{1}{\tau_a}\right)a - i\Omega_R \sigma - i\Omega_1 = 0. \quad (7)$$

We make the substitution $\tilde{a} = a + \Omega_2/\Omega_R$ and introduce the notations $\alpha = \mu_{\text{NP}}/\mu_{\text{TLS}}$, $\beta = (-i\Delta + 1/\tau_a)/\Omega - i\alpha$, $-\gamma_a = (i\Delta - 1/\tau_a)$, $-\gamma_\sigma = (i\delta - 1/\tau_\sigma)$ and $\gamma_D = 1/\tau_D$. Then, equations (5)–(7) take the form

$$2i\Omega_R(\tilde{a}^* \sigma - \sigma^* \tilde{a}) - \gamma_D(D - D_0) = 0, \quad (8)$$

$$-\gamma_\sigma \sigma + i\Omega_R \tilde{a} D = 0, \quad (9)$$

$$-\gamma_a \tilde{a} - i\Omega_R \sigma - \beta \Omega_2 = 0. \quad (10)$$

From (9) it follows that $\sigma = i\Omega_R \tilde{a} D/\gamma_\sigma$. Substituting this expression into (8) gives

$$-4\Omega_R^2 |\tilde{a}|^2 D/\gamma_\sigma - \gamma_D(D - D_0) = 0. \quad (11)$$

Finally,

$$D = D_0/[1 + 4\Omega_R^2 |\tilde{a}|^2/(\gamma_D \gamma_\sigma)], \quad (12)$$

$$\sigma = i\tilde{a} D_0 (\Omega_R/\gamma_\sigma)/[1 + 4\Omega_R^2 |\tilde{a}|^2/(\gamma_D \gamma_\sigma)], \quad (13)$$

where $\tilde{\gamma}_\sigma = |\tilde{\gamma}_\sigma|^2/\text{Re} \tilde{\gamma}_\sigma$. Now, substituting (12) and (13) into (7), after transformations we obtain

$$\beta \Omega_2 = \gamma_a \tilde{a} \left(1 - \frac{D_0 \Omega_R^2/(\gamma_a \gamma_\sigma)}{1 + 4\Omega_R^2 |\tilde{a}|^2/(\gamma_D \gamma_\sigma)}\right). \quad (14)$$

By introducing notations for the intensity of radiation acting on a spaser, $I_{\text{in}} = \Omega_2^2$, and for the intensity of the spaser response, $I_{\text{out}} = |\tilde{a}|^2$, we obtain

$$|\beta|^2 I_{\text{in}} = \gamma_a^2 I_{\text{out}} \left|1 - \frac{D_0 \Omega_R^2/(\gamma_a \gamma_\sigma)}{1 + 4\Omega_R^2 I_{\text{out}}/(\gamma_D \gamma_\sigma)}\right|^2, \quad (15)$$

or

$$I_{\text{out}} = T(I_{\text{out}}) I_{\text{in}}. \quad (16)$$

Here, by analogy with the theory of nonlinear resonator bistability [51] we introduce a nonlinear ‘transmission coefficient’, which relates the incident radiation intensity with the spaser response intensity:

$$T(I_{\text{out}}) = \frac{|\beta|^2}{\gamma_a^2} \left|1 - \frac{D_0 \Omega_R^2/(\gamma_a \gamma_\sigma)}{1 + 4\Omega_R^2 I_{\text{out}}/(\gamma_D \gamma_\sigma)}\right|^{-2}. \quad (17)$$

The dependence of T on the spaser response intensity in the case of normal initial population inversion, $D_0 < 0$, is shown in Fig. 1.

The value of the field intensity I_{out} of a nanoparticle is determined by the solution of equation (16), i.e., the intersection of the curve $T(I_{\text{out}})$ and straight line $I_{\text{out}}/I_{\text{in}}$, the slope of which depends on the external field intensity I_{in} . One can see that at certain values of I_{in} , there are three values of the field

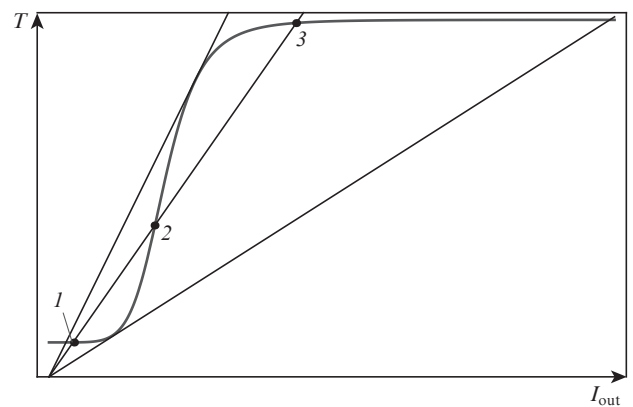


Figure 1. Dependence of the nonlinear transmission coefficient of a spaser on the spaser response intensity; points (1) and (3) show stable values of the intensity I_{out} , point (2) shows the unstable value.

intensity I_{out} (see Fig. 1). The values shown by points (1) and (3) are stable and by point (2) are unstable. Thus, there are two stable values of the field intensity of a nanoparticle, one of which is several times larger than the other. The phenomenon of bistability occurs when $D_0 \leq -8D_{\text{th}}$ [7], where $D_{\text{th}} = (1 + \Delta^2 \tau_a^2) / (\Omega_R^2 \tau_a \tau_\sigma)$ is the threshold inversion in the case of spasing of a single spaser [22, 24]. Note that bistability can appear due to the external field, even in the absence of pumping, when $D_0 = -1$.

The obtained result has a clear physical meaning. If the field of a nanoparticle is initially sufficiently large, it can cause an increase in the population inversion in a quantum dot. The external field also tends to increase the inversion in the quantum dot up to its saturation, resulting in an increase in the inversion from negative values to near zero values, which in turn leads to an increase in the field of the nanoparticle. If the field is initially small, the external field is not enough to saturate the population inversion of the quantum dot, which in this case is determined by the pump only, i.e., it is less than zero. Then, the field of the nanoparticle is still low, which explains the possibility of existence of two stable solutions.

7. Switching waves in a chain of spasers

Let us consider now the dynamics of a chain of spasers placed in an external field.

Note that an optical wave incident at some angle can excite the waves that propagate even along a chain of passive nanoparticles [17, 48, 52–54]. To ignore such waves which are not related to the bistability, we consider a scheme in which the field is perpendicular to the chain of spasers, so that the spasers are excited in-phase. This scheme is similar to that proposed in [55], where a chain of passive nanoparticles was considered.

The system of equations (2)–(4), describing the chain of interacting spasers, is as follows:

$$\begin{aligned} \dot{\hat{D}}_n &= 2i\Omega_R(\hat{a}_n^\dagger \hat{\sigma}_n - \hat{\sigma}_n^\dagger \hat{a}_n) + 2i\Omega_{\text{NP-TLS}}(\hat{a}_{n-1}^\dagger \hat{\sigma}_n - \hat{\sigma}_n^\dagger \hat{a}_{n-1}) \\ &+ 2i\Omega_{\text{NP-TLS}}(\hat{a}_{n+1}^\dagger \hat{\sigma}_n - \hat{\sigma}_n^\dagger \hat{a}_{n+1}) - \tau_D^{-1}(\hat{D}_n - \hat{D}_{0n}) \\ &+ 2i\Omega_2(\hat{\sigma}_n - \hat{\sigma}_n^\dagger), \end{aligned} \quad (18)$$

$$\begin{aligned} \dot{\hat{\sigma}} &= (i\delta - \tau_\sigma^{-1})\hat{\sigma}_n + i\Omega_R \hat{a}_n \hat{D}_n + i\Omega_{\text{NP-TLS}} \hat{a}_{n-1} \hat{D}_n \\ &+ i\Omega_{\text{NP-TLS}} \hat{a}_{n+1} \hat{D}_n + i\Omega_2 \hat{D}_n, \end{aligned} \quad (19)$$

$$\begin{aligned} \dot{\hat{a}}_n &= (i\Delta - \tau_a^{-1})\hat{a}_n - i\Omega_R \hat{\sigma}_n - i\Omega_{\text{NP-TLS}}(\hat{\sigma}_{n-1} + \hat{\sigma}_{n+1}) \\ &- i\Omega_{\text{NP-NP}}(\hat{a}_{n-1} + \hat{a}_{n+1}) - i\Omega_1, \end{aligned} \quad (20)$$

where the interaction of neighbouring spasers is described by introducing the parameters $\Omega_{\text{NP-NP}}$ and $\Omega_{\text{NP-TLS}}$, i.e., the constants of interaction of a nanoparticle with neighbouring nanoparticles and quantum dots, respectively [52].

Before we examine the dynamic regimes in a chain of spasers, we consider the solution in the form of synchronous oscillations of the chain by putting $\{a_{n-1}, \sigma_{n-1}, D_{n-1}\} = \{a_{n+1}, \sigma_{n+1}, D_{n+1}\} = \{a_n, \sigma_n, D_n\}$ in (18)–(20). This substitution, as is easily seen, reduces the problem of a chain of spasers to the problem of a single spaser with the replacement of Ω_R by $\Omega_R + 2\Omega_{\text{NP-TLS}}$ and of Δ by $\Delta - 2\Omega_{\text{NP-NP}}$ [52].

As follows from the above formulas for a single spaser for $D_0 \leq -8D_{\text{th}}$, where

$$D_{\text{th}} = \frac{1 + (\Delta - 2\Omega_{\text{NP-NP}})^2 \tau_a^2}{(\Omega_R + 2\Omega_{\text{NP-TLS}})^2 \tau_a \tau_\sigma},$$

system (18)–(20) has two stable solutions, i.e., its behaviour is bistable. However, in this case, the system consists of a chain of elements, each of which is bistable. It is well known that switching waves (concentration and temperature autowaves, population waves, etc. [56]) can propagate in distributed open nonlinear systems, in which one phase (stable) is driven out by the other (metastable).

We have studied numerically the dynamics of a chain of spasers by the FDTD method. To do this, we have selected the following initial conditions: one half of the chain is in one state and the other half is in another state. The numerical experiment has shown that a switching wave propagates in the system, the wave representing a moving interface between two stable states. The type of switching depends on the external field. In the case of a small amplitude of the external field the phase with high inversion is driven out and in the case of a high amplitude – vice versa (Fig. 2). At a certain value of the external field, which can be found numerically, the interface is at rest.

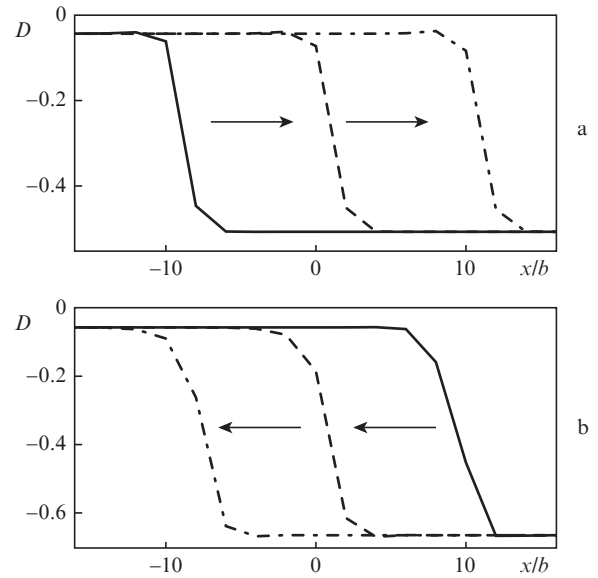


Figure 2. Switching wave propagation along the chain of spasers with a period b (direction of the front movement depends on the external field) at $\gamma_a = 10^{14} \text{ s}^{-1}$, $\gamma_\sigma = 10^{11} \text{ s}^{-1}$, $\gamma_D = 10^{13} \text{ s}^{-1}$, $\Omega_R = 10^{13} \text{ s}^{-1}$ and $D_0 = -1$ in the case of $E\mu_{\text{TLS}}/\hbar = 0.04$ (a) and 0.035 (b).

8. Formation of dissipative structures in a chain of spasers

In distributed dissipative systems, in addition to switching waves, stationary (so-called dissipative) structures can be also produced [56]. A dissipative structure represents a ‘contrast’ stationary distribution of the system parameters, inevitably arising under any initial conditions (spots on the skin of a leopard, Bernard cells, etc. [57]). In a chain of spasers such structures will be observed at losses in the nanoparticle, τ_a^{-1} , lower than in the regime of switching wave propagation.

As an initial condition we take the local perturbation of the population inversion, as shown in Fig. 3a: in the system which is one of the stable states, the nucleus of the other phase

is generated. Then, in the course of the time evolution of the chain, there will appear a dissipative structure (Fig. 3), the formation dynamics of which is of ‘self-assembly’ character. Self-assembly is a typical process of appearance of dissipative structures, when the structure self-assembles, the speed of the process and the final result being independent of the initial conditions [58, 59]. An interesting feature of self-assembly of a dissipative structure in a chain of spasers is that it appears in the same bistable regime as the switching waves.

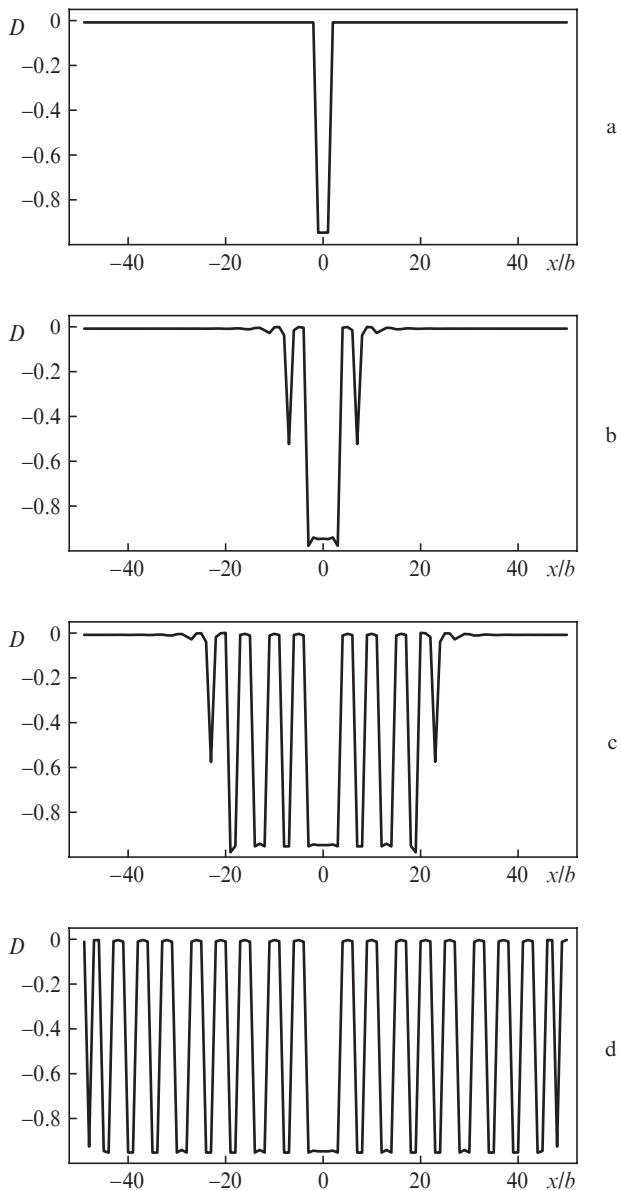


Figure 3. Formation of dissipative structures in the chain of spasers at $\gamma_a = 10^{13} \text{ s}^{-1}$, $\gamma_\sigma = 10^{11} \text{ s}^{-1}$, $\gamma_D = 10^{13} \text{ s}^{-1}$, $\Omega_R = 10^{13} \text{ s}^{-1}$ and $D_0 = -1$ in the case of $t = 0$ (a), 10^{-11} (b), 7×10^{-11} (c) and 3.5×10^{-10} s (d).

9. Conclusions

We have considered the physical mechanism and criteria of bistability of a spaser. We have shown that bistability appears at sufficiently low (below the threshold of autonomous spasing) pump levels. We have calculated the effective transmittance of the spaser allowing the ‘mirrorless’ bistability of the spaser

to be treated as the ordinary optical bistability of a nonlinear Fabry–Perot cavity [51].

Because for the development of electrodynamics of meta-materials of great interest is consideration of not a single spaser but of the whole structures made up of chains of spasers, we have investigated the effect of bistability on the dynamics coupled spasers forming a 1D chain. We have shown that such a system exhibits the behaviour typical of nonequilibrium dissipative media. In particular, at high losses in a spaser, a nonlinear switching autowave propagates in the chain, which transforms this chain from the state of low population inversion into the state of high population inversion. The control parameter, which determines the direction of the autowaves, is the amplitude of the external optical wave. At small losses there appear quasi-periodic dissipative structures, the formation dynamics of which is of ‘self-assembly’ nature.

The effect of bistability can find specific applications in plasmonics. The dipole moments of a nanoparticle and quantum dot, as well as the population inversion of the latter vary greatly near the point of bistability, which allows a spaser to be used as a transistor for optical computers. Spasers can operate as optical transistors; memory elements; pulse shapers, eliminating the noise of the incident light; discriminators; and bidirectional limiters. Another possible application of bistability is the use of a spaser to convert cw radiation into pulsed radiation. Switching waves in a bistable chain of spasers can be used to switch the spasers’ medium from a low-inverted state to a high-inverted state. Contrast dissipative structures emerging in the medium of bistable spaser can serve as masks for optical recording systems.

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