Active Plasmonics and Tuneable Plasmonic Metamaterials
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Preface

Plasmonics provides unique advantages in all areas of science and technology where the manipulation of light at the nanoscale is a prominent ingredient. It relies on light-driven coherent oscillations of electrons near a metal surface, called surface plasmons, which can trap electromagnetic waves at subwavelength and nanoscales. Thus, light can be efficiently manipulated using these plasmonic excitations. In recent years, nanostructured metals have allowed unprecedented control over optical properties and linear as well as nonlinear optical processes. As a consequence, nanoplasmonics has become a major research area and is making important advances in several main fields of applications, such as information technologies, energy, high-density data storage, life sciences, and security. In parallel, in the field of metamaterials, plasmonic composites are indispensable for achieving new optical properties in the visible and infrared spectral range, such as negative refractive index, super-resolution, and cloaking. Various types of metamaterials based on split-ring resonators, fishnet-type structures, nanorods, and others have been proposed in the whole spectral range, from microwaves to visible wavelengths. A variety of passive plasmonic elements such as mirrors, lenses, waveguides, and resonators have been demonstrated. After these initial successes, the development of active plasmonic elements capable of controlling light on the nanoscale dimensions with external electronic or optical stimuli is now on the agenda. For example, the availability of tuneable metamaterials with optical properties controlled by the external signals will immensely broaden their possible field of applications.

The number of research publications in both passive and active plasmonics and metamaterials is growing very fast. We felt that it is important to provide for readers interested in these fields a “one-stop shop” with information on the fundamentals and the state of the art in this field. Active plasmonics and metamaterials are fastmoving
fields of research and we could not possibly provide exhaustive coverage of all topics in this field. Instead, this book contains chapters from world-leading authorities in the field, covering active plasmonics from basic principles to the most recent application breakthroughs. The former covers quantum and nonlinear plasmonics, amplification of plasmonic signals and spasers, transformation optics for design of plasmonic nanostructures, active and nonlinear plasmonic metamaterials, and light control via designed phase discontinuities. The latter includes integrated plasmonic detectors, subwavelength imaging with anisotropic metamaterials, tuneable plasmonic lenses, as well as terahertz plasmonic surfaces for sensing.

The book begins with a review of gain in nanoplasmonics. This includes topics such as the spaser and plasmonic gain, amplification, and loss compensation. Both fundamental theoretical concepts and experimental developments have been reviewed. In Chapter 2, physical mechanisms responsible for nonlinearities in plasmonic nanostructures are discussed. Nonlinear surface plasmon polaritons, plasmon solitons, and nonlinear waveguide devices are presented, together with a survey of nonlinearities enhanced by localized surface plasmons and nanoantennas. Plasmonic nanorod metamaterials are introduced in Chapter 3: their optical properties as well as active and nonlinear properties allow tuneable optical responses. Transformation optics provides an extremely powerful tool for designing plasmonic nanostructures with a desired optical response, such as broadband light harvesting and nanofocusing structures, as well as cavities tailored for fluorescence enhancement in the vicinity of complex nanostructures. This new toolkit, as well as the impact of nonlocality on the optical properties of complex plasmonic nanostructures, is presented in Chapter 4.

The next chapter, building on the theoretical paradigms discussed earlier, reviews work conducted on SPP loss compensation and amplification in various types of SPP waveguides. Chapter 6 introduces the principles of light control via phase response engineering in plasmonic antennas. Generalized laws of reflection and refraction in the presence of linear interfacial phase distributions, demonstrations of giant and tuneable birefringence, and generation of optical vortices that carry orbital angular momentum are discussed. The integration of various types of nanoscale photodetectors with plasmonic waveguides and nanostructures is overviewed in Chapter 7. The adaptation of plasmonic concepts from the optical region of the spectrum to the THz band is briefly surveyed from the point of view of sensing applications in Chapter 8. The plasmonic guiding modalities in spectral range are realized either with semiconductors or via patterned metal surfaces. This chapter concludes with a description of possibilities offered by the exciting new material, graphene. A detailed review of subwavelength imaging using extremely anisotropic media in the canalization regime is presented in Chapter 9. Finally, Chapter 10 presents an overview of planar plasmonic diffractive focusing devices. The imaging properties of such plasmonic lenses can be externally tuned by controlling refractive index of the dielectric components of the nanostructure.

There are a variety of new and exciting developments outside the scope of this book, ranging from control over single-photon emission with quantum dots coupled to metallic nanostructures and the exploration of the quantum regime of SPPs, to more applied topics such as integrated nanobiosensors with optical or electrical readout. In
general, we anticipate that the combination of plasmonic structures, be it waveguides or nanoparticle cavities, with active media, will greatly accelerate the transition of plasmonics into devices. The chapters in this book provide a snapshot of the exciting possibilities that lie ahead.

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1

Spaser, Plasmonic Amplification, and Loss Compensation

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1.1 INTRODUCTION TO SPASERS AND SPASING

Not just a promise anymore [1], nanoplasmonics has delivered a number of important applications: ultrasensing [2], scanning near-field optical microscopy [3, 4], surface plasmon (SP)-enhanced photodetectors [5], thermally assisted magnetic recording [6], generation of extreme UV (EUV) [7], biomedical tests [2, 8], SP-assisted thermal cancer treatment [9], plasmonic-enhanced generation of EUV pulses [7] and extreme ultraviolet to soft x-ray (XUV) pulses [10], and many others—see also Reference 11 and 12.

To continue its vigorous development, nanoplasmonics needs an active device—near-field generator and amplifier of nanolocalized optical fields, which has until recently been absent. A nanoscale amplifier in microelectronics is the metal-oxide-semiconductor field effect transistor (MOSFET) [13, 14], which has enabled all contemporary digital electronics, including computers and communications, and the present-day technology as we know it. However, the MOSFET is limited by frequency and bandwidth to $\lesssim 100$ GHz, which is already a limiting factor in further technological development. Another limitation of the MOSFET is its high sensitivity to temperature, electric fields, and ionizing radiation, which limits its use in extreme environmental conditions and nuclear technology and warfare.

An active element of nanoplasmonics is the spaser (Surface Plasmon Amplification by Stimulated Emission of Radiation), which was proposed [15, 16] as a nanoscale quantum generator of nanolocalized coherent and intense optical fields. The idea of spaser has been further developed theoretically [17–26]. Spaser effect has recently
been observed experimentally [27]. Also a number of surface plasmon polariton (SPP) spasers (also called nanolasers) have been experimentally observed [28–33], see also References 34–37. Closely related to the spaser are nanolasers built on deep subwavelength metal nanocavities [38, 39].

1.2 SPASER FUNDAMENTALS

Spaser is a nanoplasmonic counterpart of laser [15,17]: It is a quantum generator and nanoamplifier where photons as the participating quanta are replaced by SPs. Spaser consists of a metal nanoparticle, which plays the role of a laser cavity (resonator), and the gain medium. Figure 1.1 schematically illustrates the geometry of a spaser as introduced in the original article [15], which contains a V-shaped metal nanoparticle surrounded by a layer of semiconductor nanocrystal quantum dots (QDs).

The laser has two principal elements: resonator (or cavity) that supports photonic mode(s) and the gain (or active) medium that is population-inverted and supplies energy to the lasing mode(s). An inherent limitation of the laser is that the size of the laser cavity in the propagation direction is at least half the wavelength and practically more than that even for the smallest lasers developed [28, 29, 40].

In a true spaser [15, 18], this limitation is overcome. The spasing modes are SPs whose localization length is on the nanoscale [41] and is only limited by the minimum inhomogeneity scale of the plasmonic metal and the nonlocality radius [42] $l_{nl} \sim 1 \text{ nm}$. This nonlocality length is the distance that an electron with the Fermi velocity $v_F$ moves in space during a characteristic period of the optical field:

$$l_{nl} \sim v_F / \omega \sim 1 \text{ nm}, \quad (1.1)$$

where $\omega$ is the optical frequency, and the estimate is shown for the optical spectral region. So, the spaser is truly nanoscopic—its minimum total size can be just a few nanometers.

![FIGURE 1.1](image_url)  
**FIGURE 1.1** Schematic of the spaser as originally proposed in Reference 15. The resonator of the spaser is a metal nanoparticle shown as a gold V-shape. It is covered by the gain medium depicted as nanocrystal quantum dots. This active medium is supported by a neutral substrate.
The resonator of a spaser can be any plasmonic metal nanoparticle whose total size $R$ is much less than the wavelength $\lambda$ and whose metal thickness is between $l_{sl}$ and $l_s$, which supports an SP mode with required frequency $\omega_n$. Here $l_s$ is the skin depth:

$$l_s = \tilde{\lambda} \left[ \text{Re} \left( \frac{\epsilon_m^2}{\epsilon_m + \epsilon_d} \right)^{1/2} \right]^{-1}, \quad (1.2)$$

where $\tilde{\lambda} = \lambda/(2\pi) = \omega/c$ is the reduced vacuum wavelength, $\epsilon_m$ is the dielectric function (or, permittivity) of the metal, and $\epsilon_d$ is that of the embedding dielectric. For single-valence plasmonic metals (silver, gold, copper, alkaline metals) $l_s \approx 25$ nm in the entire optical region.

This metal nanoparticle should be surrounded by the gain medium that overlaps with the spasing SP eigenmode spatially and whose emission line overlaps with this eigenmode spectrally [15]. As an example, we consider in more detail a model of a nanoshell spaser [17, 18, 43], which is illustrated in Figure 1.2. Panel (a) shows a silver nanoshell carrying a single SP (plasmon population number $N_n = 1$) in the dipole eigenmode. It is characterized by a uniform field inside the core and hot spots at the poles outside the shell with the maximum field reaching $\sim 10^6$ V/cm. Similarly, Figure 1.2b shows the quadrupole mode in the same nanoshell. In this case, the mode electric field is nonuniform, exhibiting hot spots of $\sim 1.5 \times 10^6$ V/cm of the modal electric field at the poles. These high values of the modal fields, which are related to the small modal volume, are the underlying physical reason for a very strong feedback in the spaser. Under our conditions, the electromagnetic retardation within the spaser volume can be safely neglected. Also, the radiation of such a spaser is a weak effect: The decay rate of plasmonic eigenmodes is dominated by the internal loss in the metal. Therefore, it is sufficient to consider only quasistatic eigenmodes [41, 44] and not their full electrodynamic counterparts [45].

For the sake of numerical illustrations of our theory, we will use the dipole eigenmode (Fig. 1.2a). There are two basic ways to place the gain medium: (i) outside the nanoshell, as shown in panel (c), and (ii) in the core, as in panel (d), which was originally proposed in Reference 43. As we have verified, these two designs lead to comparable characteristics of the spaser. However, the placement of the gain medium inside the core illustrated in Figure 1.2d has a significant advantage because the hot spots of the local field are not covered by the gain medium and are sterically available for applications.

Note that any $l$-multipole mode of a spherical particle is, indeed, $2l + 1$-times degenerate. This may make the spasing mode to be polarization unstable, like in lasers without polarizing elements. In reality, the polarization may be clamped and become stable due to deviations from the perfect spherical symmetry, which exist naturally or can be introduced deliberately. More practical shape for a spaser may be a nanorod [24], which has a mode with the stable polarization along the major axis. However, a nanorod is a more complicated geometry for theoretical treatment.

The level diagram of the spaser gain medium and the plasmonic metal nanoparticle is displayed in Figure 1.2e along with a schematic of the relevant energy transitions in
FIGURE 1.2  Schematic of spaser geometry, local fields, and fundamental processes leading to spasing. (a) Nanoshell geometry and the local optical field distribution for one SP in an axially symmetric dipole mode. The nanoshell has aspect ratio \( \eta = 0.95 \). The local-field magnitude is color-coded by the scale bar in the right-hand side of the panel. (b) The same as (a) but for a quadrupole mode. (c) Schematic of a nanoshell spaser where the gain medium is outside of the shell, on the background of the dipole-mode field. (d) The same as (c) but for the gain medium inside the shell. (e) Schematic of the spasing process. The gain medium is excited and population-inverted by an external source, as depicted by the black arrow, which produces electron–hole pairs in it. These pairs relax, as shown by the green arrow, to form the excitons. The excitons undergo decay to the ground state emitting SPs into the nanoshell. The plasmonic oscillations of the nanoshell stimulate this emission, supplying the feedback for the spaser action. Adapted from Reference 18.
the system. The gain medium chromophores may be semiconductor nanocrystal QDs [15, 46], dye molecules [47, 48], rare-earth ions [43], or electron–hole excitations of an unstructured semiconductor [28, 40]. For certainty, we will use a semiconductor-science language of electrons and holes in QDs.

The pump excites electron–hole pairs in the chromophores (Fig. 1.2e), as indicated by the vertical black arrow, which relax to form excitons. The excitons constitute the two-level systems that are the donors of energy for the SP emission into the spasing mode. In vacuum, the excitons would recombine emitting photons. However, in the spaser geometry, the photoemission is strongly quenched due to the resonance energy transfer to the SP modes, as indicated by the red arrows in the panel. The probability of the radiativeless energy transfer to the SPs relative to that of the radiative decay (photon emission) is given by the so-called Purcell factor

$$\sim \frac{\lambda^3 Q}{R^3} \gg 1,$$

where $R$ is a characteristic size of the spaser metal core and $Q$ is the plasmonic quality factor [12], and $Q \sim 100$ for a good plasmonic metal such as silver. Thus, this radiativeless energy transfer to the spaser mode is the dominant process whose probability is by orders of magnitude greater than that of the free-space (far-field) emission.

The plasmons already in the spaser mode create the high local fields that excite the gain medium and stimulate more emission to this mode, which is the feedback mechanism. If this feedback is strong enough, and the lifetime of the spaser SP mode is long enough, then an instability develops leading to the avalanche of the SP emission in the spasing mode and spontaneous symmetry breaking, establishing the phase coherence of the spasing state. Thus the establishment of spasing is a nonequilibrium phase transition, as in the physics of lasers.

1.2.1 Brief Overview of the Latest Progress in Spasers

After the original theoretical proposal and prediction of the spaser [15], there has been an active development in this field, both theoretical [17–26] and experimental [27–33]; see also [11, 12]. There has also been a US patent issued on spaser [16].

Among theoretical developments, a nanolens spaser has been proposed [49], which possesses a nanofocus (“the hottest spot”) of the local fields. In References 15 and 49, the necessary condition of spasing has been established on the basis of the perturbation theory.

There have been theories published describing the SPP spasers (or “nanolasers” as sometimes they are called) phenomenologically, on the basis of classic linear electrodynamics by considering the gain medium as a dielectric with a negative imaginary part of the permittivity (e.g., [43]). Very close fundamentally and technically are works on the loss compensation in metamaterials [50–53]. Such linear-response approaches do not take into account the nature of the spasing as a nonequilibrium phase transition, at the foundation of which is spontaneous symmetry breaking: establishing coherence.
Spaser, Plasmonic Amplification, and Loss Compensation

with an arbitrary but sustained phase of the SP quanta in the system [18]. Spaser is necessarily a deeply nonlinear (nonperturbative) phenomenon where the coherent SP field always saturates the gain medium, which eventually brings about establishment of the stationary (or continuous wave (CW)) regime of the spasing [18]. This leads to principal differences of the linear-response results from the microscopic quantum-mechanical theory in the region of spasing, as we discuss below in conjunction with Figure 1.4.

There has also been a theoretical publication on a bow tie spaser (nanolaser) with electrical pumping [54]. It is based on balance equations and only the CW spasing generation intensity is described. Yet another theoretical development has been a proposal of the lasing spaser [55], which is made of a plane array of spasers.

There has also been a theoretical proposal of a spaser (“nanolaser”) consisting of a metal nanoparticle coupled to a single chromophore [56]. In this paper, a dipole–dipole interaction is illegitimately used at very small distances $r$ where it has a singularity (diverging for $r \to 0$), leading to a dramatically overestimated coupling with the SP mode. As a result, a completely unphysical prediction of CW spasing due to single chromophore has been obtained [56]. In contrast, our theory [18] is based on the full (exact) field of the spasing SP mode without the dipole (or any multipole) approximation. As our results of Section 1.3.4 below show, hundreds of chromophores per metal nanoparticle are realistically required for the spasing even under the most favorable conditions.

There has been a vigorous experimental investigation of the spaser and the concepts of spaser. Stimulated emission of SPPs has been observed in a proof-of-principle experiment using pumped dye molecules as an active (gain) medium [47]. There have also been later experiments that demonstrated strong stimulated compensating a significant part of the SPP loss [48, 57–61]. As a step toward the lasing spaser, the first experimental demonstration has been reported of a partial compensation of the Joule losses in a metallic photonic metamaterial using optically pumped PbS semiconductor QDs [46]. There have also been experimental investigations reporting the stimulated emission effects of SPs in plasmonic metal nanoparticles surrounded by gain media with dye molecules [62, 63]. The full loss compensation and amplification of the long-range SPPs at $\lambda = 882$ nm in a gold nanostrip waveguide with a dye solution as a gain medium has been observed [64].

At the present time, there have been a considerable number of successful experimental observations of spasers and SPP spasers (also called nanolasers). An electrically pumped nanolaser with semiconductor gain medium has been demonstrated [28] where the lasing modes are SPPs with a one-dimensional (1d) confinement to a $\sim 50$ nm size. A nanolaser with an optically pumped semiconductor gain medium and a hybrid semiconductor/metal (CdS/Ag) SPP waveguide has been demonstrated with an extremely tight transverse (2d) mode confinement to $\sim 10$ nm size [29]. This has been followed by the development of a CdS/Ag nanolaser generating a visible single mode at room temperature with a tight 1d confinement ($\sim 20$ nm) and a 2d confinement in the plane of the structure to an area $\sim 1 \mu m^2$ [30]. A highly efficient SPP spaser in the communication range ($\lambda = 1.46 \mu m$) with an optical pumping based on a gold film and an InGaAs semiconductor quantum-well gain medium has recently been reported [31]. Another nanolaser (spaser) has been reported based on
gold as a plasmonic metal and InGaN/GaN nanorods as gain medium [32]. This spaser generates in the green optical range. Also a promising type of spasers has been introduced [33] based on distributed feedback (DFB). The nanolaser demonstrated in Reference 33 generates at room temperature and has lower threshold than other spasers—see also the corresponding discussion in Section 1.4.6.

There has been an observation published of a nanoparticle spaser [27]. This spaser is a chemically synthesized gold nanosphere of radius 7 nm surrounded by a dielectric shell of 21 nm outer radius containing immobilized dye molecules. Under nanosecond optical pumping in the absorption band of the dye, this spaser develops a relatively narrow-spectrum and intense visible emission that exhibits a pronounced threshold in pumping intensity. The observed characteristics of this spaser are in an excellent qualitative agreement and can be fully understood on the basis of the corresponding theoretical results described in Section 1.3.4.

1.3 QUANTUM THEORY OF SPASER

1.3.1 Surface Plasmon Eigenmodes and Their Quantization

Here we will follow References 41, 65, and 66 to introduce SPs as eigenmodes and Reference 15 to quantize them. Assuming that a nanoplasmonic system is small enough, \( R \ll \lambda, R \lesssim l_s \), we employ the so-called quasistatic approximation where the Maxwell equations reduce to the continuity equation for the electrostatic potential \( \psi(r) \):

\[
\frac{\partial}{\partial r} \varepsilon(r) \frac{\partial}{\partial r} \psi(r) = 0. \tag{1.4}
\]

The systems permittivity (dielectric function) varying in space and frequency-dependent is expressed as

\[
\varepsilon(r, \omega) = \varepsilon_m(\omega) \Theta(r) + \varepsilon_d[1 - \Theta(r)] = \varepsilon_d \left[ 1 - \frac{\Theta(r)}{s(\omega)} \right]. \tag{1.5}
\]

Here \( \Theta(r) \) is the so-called characteristic function of the nanosystem, which is equal to 1 when \( r \) belongs to the metal and 0 otherwise. We have also introduced Bergman’s spectral parameter [44]:

\[
s(\omega) = \frac{\varepsilon_d}{\varepsilon_d - \varepsilon_m(\omega)}. \tag{1.6}
\]

A classical-field SP eigenmode \( \psi_n(r) \) is defined by the following generalized eigenproblem, which is obtained from Equation (1.4) by substituting Equations (1.5) and (1.6):

\[
\frac{\partial}{\partial r} \Theta(r) \frac{\partial}{\partial r} \psi_n(r) - s_n \frac{\partial^2}{\partial r^2} \psi_n(r) = 0, \tag{1.7}
\]
where $\omega_n$ is the corresponding eigenfrequency and $s_n = s(\omega_n)$ is the corresponding eigenvalue.

To be able to carry out the quantization procedure, we must neglect losses, that is, consider a purely Hamiltonian system. This requires that we neglect $\text{Im} \varepsilon_m$, which we do only in this subsection. Then the eigenvalues $s_n$ and the corresponding SP wave functions $\varphi_n$, as defined by Equation (1.7), are all real. Note that for good metals in the plasmonic region, $\text{Im} \varepsilon_m \ll |\text{Re} \varepsilon_m|$, cf. Reference 12, so this procedure is meaningful.

The eigenfunctions $\varphi_n(\mathbf{r})$ satisfy the homogeneous Dirichlet–Neumann boundary conditions on a surface $S$ surrounding the system. These we set as

$$ \varphi_1(\mathbf{r})|_{\mathbf{r} \in S} = 0, \text{ or } \mathbf{n}(\mathbf{r}) \frac{\partial}{\partial \mathbf{r}} \varphi_1(\mathbf{r}) \bigg|_{\mathbf{r} \in S} = 0, \quad (1.8) $$

with $\mathbf{n}(\mathbf{r})$ denoting a normal to the surface $S$ at a point of $\mathbf{r}$.

From Equations (1.4), (1.5), (1.6), (1.7), and (1.8) it is straightforward to obtain that

$$ \int_V \varepsilon(\mathbf{r}, \omega) |\nabla \varphi_n(\mathbf{r})|^2 dV = \varepsilon_d \left[ 1 - \frac{s_n}{s(\omega)} \right], \quad (1.9) $$

where $V$ is the volume of the system.

To quantize the SPs, we write the operator of the electric field of an SP eigenmode as a sum over the eigenmodes:

$$ \hat{\mathbf{E}}(\mathbf{r}) = -\sum_n A_n \nabla \varphi_n(\mathbf{r})(\hat{a}_n + \hat{a}_n^\dagger), \quad (1.10) $$

where $\hat{a}_n^\dagger$ and $\hat{a}_n$ are the SP creation and annihilation operators, $-\nabla \varphi_n(\mathbf{r}) = \mathbf{E}_n(\mathbf{r})$ is the modal field of an $n$th mode, and $A_n$ is an unknown normalization constant. Note that $\hat{a}_n^\dagger$ and $\hat{a}_n$ satisfy the Bose–Einstein canonical commutation relations,

$$ [\hat{a}_n, \hat{a}_m^\dagger] = \delta_{mn}, \quad (1.11) $$

where $\delta_{mn}$ is the Kronecker symbol.

To find normalization constant $A_n$, we invoke Brillouin’s expression [67] for the average energy $\langle \hat{H}_{SP} \rangle$ of SPs as a frequency-dispersive system:

$$ \langle \hat{H}_{SP} \rangle = \frac{1}{8\pi} \int_V \frac{\partial}{\partial \omega} \left[ \omega \varepsilon(\mathbf{r}, \omega) \right] \sum_n (\hat{\mathbf{E}}_n^\dagger(\mathbf{r}) \hat{\mathbf{E}}_n(\mathbf{r})) dV, \quad (1.12) $$

where

$$ \hat{H}_{SP} = \sum_n \hbar \omega_n \left( \hat{a}_n^\dagger \hat{a}_n + \frac{1}{2} \right) \quad (1.13) $$

is the SP Hamiltonian in the second quantization.
Finally, we substitute the field expansion (1.10) into Equation (1.12) and take into account Equation (1.9) to carry out the integration. Comparing the result with Equation (1.13), we immediately obtain an expression for the quantization constant:

\[
A_n = \left( \frac{4\pi \hbar s'_n}{\varepsilon d s'_n} \right)^{1/2}, \quad s'_n \equiv \text{Re} \frac{ds(\omega)}{d\omega} \bigg|_{\omega = \omega_n}
\]  

(1.14)

Note that we have corrected a misprint in Reference 15 by replacing the coefficient \(2\pi\) by \(4\pi\).

1.3.2 Quantum Density Matrix Equations (Optical Bloch Equations) for Spaser

Here we follow Reference 18. The spaser Hamiltonian has the form

\[
\hat{H} = \hat{H}_g + \hat{H}_{SP} - \sum_p \hat{E}(r_p) \hat{d}(p),
\]

(1.15)

where \(\hat{H}_g\) is the Hamiltonian of the gain medium, \(p\) is a number (label) of a gain medium chromophore, \(r_p\) is its coordinate vector, and \(\hat{d}(p)\) is its dipole-moment operator. In this theory, we treat the gain medium quantum mechanically but the SPs quasi-classically, considering \(\hat{a}_n\) as a classical quantity (c-number) \(a_n\) with time dependence as \(a_n = a_{0n} \exp(-i\omega t)\), where \(a_{0n}\) is a slowly varying amplitude. The number of coherent SPs per spasing mode is then given by \(N_p = |a_{0n}|^2\). This approximation neglects quantum fluctuations of the SP amplitudes. However, when necessary, we will take into account these quantum fluctuations, in particular, to describe the spectrum of the spaser.

Introducing \(\rho^{(p)}\) as the density matrix of a \(p\)th chromophore, we can find its equation of motion in a conventional way by commutating it with the Hamiltonian (1.15) as

\[
i\hbar \dot{\rho}^{(p)} = [\rho^{(p)}, \hat{H}],
\]

(1.16)

where the dot denotes temporal derivative. We use the standard rotating wave approximation (RWA), which only takes into account the resonant interaction between the optical field and chromophores. We denote |1\rangle and |2\rangle as the ground and excited states of a chromophore, with the transition |2\rangle \rightleftharpoons |1\rangle resonant to the spasing plasmon mode \(n\). In this approximation, the time dependence of the nondiagonal elements of the density matrix is \((\rho^{(p)})_{12} = \tilde{\rho}^{(p)}_{12} \exp(i\omega t)\) and \((\rho^{(p)})_{21} = \tilde{\rho}^{(p)*}_{12} \exp(-i\omega t)\), where \(\tilde{\rho}^{(p)}_{12}\) is an amplitude slowly varying in time, which defines the coherence (polarization) for the |2\rangle \rightleftharpoons |1\rangle spasing transition in a \(p\)th chromophore of the gain medium.

Introducing a rate constant \(\Gamma_{12}\) to describe the polarization relaxation and a difference \(n^{(p)}_{21} = \rho^{(p)}_{22} - \rho^{(p)}_{11}\) as the population inversion for this spasing transition, we derive an equation of motion for the nondiagonal element of the density matrix as

\[
\dot{\tilde{\rho}}^{(p)}_{12} = -[i(\omega - \omega_{12}) + \Gamma_{12}] \tilde{\rho}^{(p)}_{12} + i a_{0n} n^{(p)}_{21} \tilde{\Sigma}^{(p)*}_{12},
\]

(1.17)
where

\[ \tilde{\Omega}_{12}^{(p)} = -A_n \mathbf{d}_{12}^{(p)} \nabla \varphi_n(\mathbf{r}_p) / \hbar \]  

(1.18)

is the one-plasmon Rabi frequency for the spasing transition in a \( p \)th chromophore and \( \mathbf{d}_{12}^{(p)} \) is the corresponding transitional dipole element. Note that always \( \mathbf{d}_{12}^{(p)} \) is either real or can be made real by a proper choice of the quantum state phases, making the Rabi frequency \( \tilde{\Omega}_{12}^{(p)} \) also a real quantity.

An equation of motion for \( n_{21}^{(p)} \) can be found in a standard way by commutating it with \( \hat{H} \) and adding the corresponding decay and excitation rates. To provide conditions for the population inversion \( (n_{21}^{(p)} > 0) \), we imply existence of a third level. For simplicity, we assume that it very rapidly decays into the excited state \(|2\rangle\) of the chromophore, so its own population is negligible. It is pumped by an external source from the ground state (optically or electrically) with some rate that we will denote \( g \).

In this way, we obtain the following equation of motion:

\[ \dot{n}_{21}^{(p)} = -4 \text{Im} \left[ a_{0n} \tilde{\rho}_{12}^{(p)} \tilde{\Omega}_{21}^{(p)} \right] - \gamma_2 \left( 1 + n_{21}^{(p)} \right) + g \left( 1 - n_{21}^{(p)} \right), \]  

(1.19)

where \( \gamma_2 \) is the decay rate \( |2\rangle \rightarrow |1\rangle \).

The stimulated emission of the SPs is described as their excitation by the local field created by the coherent polarization of the gain medium. The corresponding equation of motion can be obtained using Hamiltonian (1.15) and adding the SP relaxation with a rate of \( \gamma_n \) as

\[ \dot{a}_{0n} = \left[ i (\omega - \omega_n) - \gamma_n \right] a_{0n} + i a_{0n} \sum_p \rho_{12}^{(p)*} \tilde{\Omega}_{12}^{(p)} \].  

(1.20)

As an important general remark, the system of Equations (1.17), (1.19), and (1.20) is highly nonlinear: Each of these equations contains a quadratic nonlinearity: a product of the plasmon-field amplitude \( a_{0n} \) by the density matrix element \( \rho_{12} \) or population inversion \( n_{21} \). Altogether, this is a six-order nonlinearity. This nonlinearity is a fundamental property of the spaser equations, which makes the spaser generation always a fundamentally nonlinear process. This process involves a nonequilibrium phase transition and a spontaneous symmetry breaking: establishment of an arbitrary but sustained phase of the coherent SP oscillations.

A relevant process is spontaneous emission of SPs by a chromophore into a spasing SP mode. The corresponding rate \( \gamma_2^{(p)} \) for a chromophore at a point \( \mathbf{r}_p \), can be found in a standard way using the quantized field (1.10) as

\[ \gamma_2^{(p)} = 2 \frac{A_n^2}{\hbar \gamma_n} \left| \mathbf{d}_{12} \nabla \varphi_n(\mathbf{r}_p) \right|^2 \frac{(\Gamma_{12} + \gamma_n)^2}{(\omega_{12} - \omega_n)^2 + (\Gamma_{12} + \gamma_n)^2}. \]  

(1.21)