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Loss compensation by gain and spasing

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We present a theory of the effective dielectric response of metamaterials containing gain. We demonstrate analytically that the conditions of spaser generation and the full loss compensation in a dense resonant plasmonic-gain metamaterial are identical. Consequently, attempting the full compensation or overcompensation of losses by gain will lead to an instability and a transition to a spaser state. This will limit (clamp) the inversion and eliminate the net gain. As a result, the full loss compensation (overcompensation) in such metamaterials is impossible. The criterion of the loss overcompensation, leading to the instability and spasing, is given in an analytical and a universal (independent from system geometry) form. Comparison with existing experiments is carried out.

Keywords: spaser; gain; amplification; nanoplasmonics; metamaterials

1. Introduction

Nanoplasmonics studies and employs optical fields that are localized on the nanoscale in the vicinity of metal nanostructures [1]. It is presently a thriving, active, and very rapidly developing field with a number of exciting physical effects and many practical applications based on the unique nanolocalization properties of plasmonic fields [2,3].

A problem for many applications of plasmonics is posed by losses inherent in the interaction of light with metals. There are several ways to bypass, mitigate or overcome the detrimental effects of these losses, which we briefly discuss below.

- The most common approach consists of employing effects where the losses are not fundamentally important such as surface plasmon polariton (SPP) propagation used in sensing [2], ultramicroscopy [4,5] and solar energy conversion [6]. For realistic losses, there are other effects and applications that are pronounced and useful, in particular, sensing based on surface plasmon (SP) resonances and surface-enhanced Raman scattering [2,7–10].
- Another promising idea is to use superconducting plasmonics to dramatically reduce losses [11–14]. However, this is only applicable for frequencies below the superconducting gap, i.e. in the terahertz region.

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- Yet another promising direction is using highly doped semiconductors where the ohmic losses can be significantly lower owing to much lower free carrier concentrations [15]. However, a problem with this approach may lie in the fact that the usefulness of plasmonic modes depends not on the loss *per se* but on the quality factor Q , which for doped semiconductors may not be higher than for the plasmonic metals.
- One of the promising alternative approaches to low-loss plasmonic metamaterials is using gain to compensate the dielectric (ohmic) losses [3,16]. In this case, the gain medium is included into the metamaterials. It surrounds the metal plasmonic component in the same manner as in spasers (see below). The idea is that the gain will provide quantum amplification compensating the loss in the metamaterial.

This paper is devoted to the theory of the loss compensation in plasmonic metamaterials using gain. The principles of the loss compensation are based on the idea of the spaser [17–19]. The spaser (SP amplification by stimulated emission of radiation) has been introduced in Bergman & Stockman [17] as a nanoplasmonic counterpart of the laser.

The spaser is built of a metal nanoplasmonic nanoparticle, which plays the role of the resonator ('cavity') of the laser, and a nanoscale gain medium (dye molecules, quantum dots (QDs), etc.) where the population inversion is created optically or electrically. When the spaser's gain medium is excited (optically or electrically), radiationless transfer of energy from the electron transitions in the gain medium to SP modes of the metal nanoparticle takes place. The local field of the SPs periodically perturbs the gain medium causing the stimulated emission of further SPs into the same mode. This is the mechanism of the quantum amplification in the spaser.

If this spaser amplification overcomes the loss in its metal core, the initial state of the system loses its stability, and a new, spasing state appears with a coherent SP population whose phase is established owing to a spontaneous symmetry breaking [20]. The spaser is a nanoscopic generator of coherent local optical fields and their ultrafast nanoamplifier. The amplification principle of the spaser can be used to compensate the dielectric and other losses in metamaterials.

After the spaser was introduced, its ideas and implementations have been very actively developed experimentally. A true nanoscopic spaser consisting of a gold nanosphere core surrounded by a dielectric gain shell containing a laser dye has been demonstrated [21].

Apart from the spaser based on nanolocalized SPs, there also are known the so-called nanolasers. A nanolaser is actually a SPP-based spaser. Such a nanolaser is based on the amplification of the propagating SPPs owing to their stimulated emission by the gain medium adjacent to the SPP waveguide. The nanolasers (SPP spasers) have been demonstrated with one-dimensional [22], two-dimensional [23] and three-dimensional [24] confinements. The latter actually differs from the original SP spaser only by the size of its eigenmodes, which is presently of the order of a half wavelength (approx. $\lambda/2$) laterally and deep nanoscopic transversely [24]. This nanolaser is able to generate in a single mode at ambient temperature.

Different from the SPP spasers, the so-called lasing spaser has been introduced [25]. This is a nanofilm (planar metamaterial also known as a metasurface) containing plasmonic resonators and the gain medium. The pre-generation narrowing of the resonant line in the lasing spaser has been demonstrated [26].

Amplification of long-range SPPs in a gold strip waveguide in the proximity of a pumped dye solution has been demonstrated [27]. Amplified spontaneous emission of SPPs has been observed in gold nanofilms over an amplifying medium containing PbS QDs, where the reduction of the SPP propagation loss by up to 30 per cent has taken place [28]. In a metamaterial consisting of split-ring resonators coupled with an optically pumped InGaAs quantum well, a reduction of the transmission loss by approximately 8 per cent has been observed [29]. The full compensation and overcompensation of the optical transmission loss for a fishnet metamaterial containing a pumped dye dispersed in a polymer matrix have been described [30]. This experiment was later stated to be in agreement with a theory based on the Maxwell–Bloch equations [31].

In this paper, we show that the full compensation or overcompensation of the optical loss in a dense resonant gain metamaterial leads to an instability that is resolved by its spasing (i.e. by becoming a spaser). We further show analytically that the conditions of the complete gain compensation and the threshold condition of spasing [20] are identical. Thus, the full compensation (overcompensation) of the loss by gain in such a metamaterial will cause spasing. This spasing limits (clamps) the gain [20] and, consequently, inhibits the complete loss compensation (overcompensation) at any frequency. Partially, a brief version of this theory, without the derivations and most details and discussions presented in this paper, has been published previously [32,33].

2. Permittivity of nanoplasmonic metamaterial

We will consider, for certainty, an isotropic and uniform metamaterial (which is defined as large with respect to the wavelength, a periodic system—plasmonic crystal—whose unit cell is much smaller than the reduced wavelength of the propagating electromagnetic waves) that, by definition, in a range of frequencies ω can be described by the effective permittivity $\bar{\epsilon}(\omega)$ and permeability $\bar{\mu}(\omega)$. We will concentrate below on the loss compensation for the optical electric responses; similar consideration with identical conclusions for the optical magnetic responses is straightforward. Consider a small piece of the metamaterial with size much greater than the unit cell but much smaller than the reduced wavelength λ , which is a metamaterial itself. Let us subject this metamaterial to a uniform electric field $\mathbf{E}(\omega) = -\nabla\phi(\mathbf{r}, \omega)$ oscillating with frequency ω . Note that $\mathbf{E}(\omega)$ is the amplitude of the macroscopic electric field inside the metamaterial. We will denote the local field at a point \mathbf{r} inside this metamaterial as $\mathbf{e}(\mathbf{r}, \omega) = -\nabla\varphi(\mathbf{r}, \omega)$. We assume standard boundary conditions

$$\varphi(\mathbf{r}, \omega) = \phi(\mathbf{r}, \omega), \quad (2.1)$$

for \mathbf{r} belonging to the surface S of the volume under consideration.

To present our results in a closed form, we first derive a homogenization formula used in Stockman *et al.* [34] (see also references cited therein). By definition, the electric displacement in the volume V of the metamaterial is given by a formula

$$\mathbf{D}(\mathbf{r}, \omega) = \frac{1}{V} \int_V \varepsilon(\mathbf{r}, \omega) \mathbf{e}(\mathbf{r}, \omega) \, dV, \quad (2.2)$$

where $\varepsilon(\mathbf{r}, \omega)$ is a position-dependent permittivity. This can be identically expressed (by multiplying and dividing by the macroscopic field E^*) and, using the Gauss theorem, transformed to a surface integral as

$$\begin{aligned} D &= \frac{1}{VE^*(\omega)} \int_V \mathbf{E}^*(\omega) \varepsilon(\mathbf{r}, \omega) \mathbf{e}(\mathbf{r}, \omega) \, dV \\ &= \frac{1}{VE^*(\omega)} \int_S \phi^*(\mathbf{r}, \omega) \varepsilon(\mathbf{r}, \omega) \mathbf{e}(\mathbf{r}, \omega) \, d\mathbf{S}, \end{aligned} \quad (2.3)$$

where we took into account Maxwell equation $\nabla[\varepsilon(\mathbf{r}, \omega) \mathbf{e}(\mathbf{r}, \omega)] = 0$. Now, using the boundary conditions of equation (2.1), we can express it and transform back to the surface integral as

$$\begin{aligned} D &= \frac{1}{VE^*(\omega)} \int_S \varphi^*(\mathbf{r}) \varepsilon(\mathbf{r}, \omega) \mathbf{e}(\mathbf{r}, \omega) \, d\mathbf{S} \\ &= \frac{1}{VE^*(\omega)} \int_V \varepsilon(\mathbf{r}, \omega) |\mathbf{e}(\mathbf{r}, \omega)|^2 \, dV. \end{aligned} \quad (2.4)$$

From the last equality, we obtain the required homogenization formula as an expression for the effective permittivity of the metamaterial:

$$\bar{\varepsilon}(\omega) = \frac{1}{V|E(\omega)|^2} \int_V \varepsilon(\mathbf{r}, \omega) |\mathbf{e}(\mathbf{r}, \omega)|^2 \, dV. \quad (2.5)$$

3. Plasmonic eigenmodes and effective resonant permittivity of metamaterial

Here, for the sake of convenience, we briefly reiterate the quasi-static spectral theory of Stockman *et al.* [35–37]. The quasi-static eigenmode equation is

$$\nabla[\theta(\mathbf{r}) \nabla \varphi_n(\mathbf{r})] = s_n \nabla^2 \varphi_n(\mathbf{r}), \quad (3.1)$$

where $\varphi_n(\mathbf{r})$ is an eigenfunction, s_n is the corresponding eigenvalue and $\theta(\mathbf{r})$ is the characteristic function that is equal to 1 inside the metal and 0 otherwise. The homogeneous Dirichlet–Neumann boundary conditions are implied.

From equation (3.1) one can easily find a relation

$$s_n = \frac{\int_V \theta(\mathbf{r}) |\mathbf{E}_n(\mathbf{r})|^2 \, dV}{\int_V |\mathbf{E}_n(\mathbf{r})|^2 \, dV}, \quad (3.2)$$

where $\mathbf{E}_n(\mathbf{r}) = -\nabla \varphi_n(\mathbf{r})$ is the eigenmode's field. We will assume that this eigenmode field is normalized according to

$$\int_V |\mathbf{E}_n(\mathbf{r})|^2 \, dV = 1. \quad (3.3)$$

This condition is arbitrary but standard and convenient.

From equation (3.1) it follows, in particular, that

$$1 \geq s_n \geq 0. \quad (3.4)$$

The resonant frequency, $\omega = \omega_n$, is defined by the equality

$$s_n = \text{Re } s(\omega_n), \quad s(\omega) \equiv \frac{\varepsilon_h(\omega)}{\varepsilon_h(\omega) - \varepsilon_m(\omega)}, \quad (3.5)$$

where $s(\omega)$ is Bergman's spectral parameter, $\varepsilon_m(\omega)$ is the permittivity of the metal and $\varepsilon_h(\omega)$ is that of the surrounding host with the gain chromophore centres.

The local field inside the nanostructured volume V of the metamaterial is given by the eigenmode expansion

$$\left. \begin{aligned} \mathbf{e}(\mathbf{r}, \omega) &= \mathbf{E}(\omega) - \sum_n \frac{a_n}{s(\omega) - s_n} \mathbf{E}_n(\mathbf{r}) \\ a_n &= \mathbf{E}(\omega) \int_V \theta(\mathbf{r}) \mathbf{E}_n(\mathbf{r}) \, dV. \end{aligned} \right\} \quad (3.6)$$

where

In the resonance, $\omega = \omega_n$, only one term in equation (3.6) dominates, and it becomes

$$\mathbf{e}(\mathbf{r}, \omega) = \mathbf{E}(\omega) + i \frac{a_n}{\text{Im } s(\omega_n)} \mathbf{E}_n(\mathbf{r}). \quad (3.7)$$

The first term in this equation corresponds to the mean (macroscopic) field and the second one describes the deviations of the local field from the mean field containing contributions of the hot spots [38], etc. The mean root square ratio of the second term (local field) to the first (mean field) is estimated as

$$\sim \frac{f}{\text{Im } s(\omega_n)} = \frac{fQ}{s_n(1 - s_n)}, \quad (3.8)$$

where we took into account that, in accord with equation (3.3), $E_n \sim V^{-1/2}$, and

$$f = \frac{1}{V} \int_V \theta(\mathbf{r}) \, dV \quad \text{and} \quad Q = -\frac{\text{Re } \varepsilon_m(\omega)}{\text{Im } \varepsilon_m(\omega)}; \quad (3.9)$$

f is the metal fill factor of the system, and Q is the plasmonic quality factor. Deriving expression (3.8), we have also taken into account an equality $\text{Im } s(\omega_n) = s_n(1 - s_n)/Q$, which is valid in the assumed limit of a high quality factor, $Q \gg 1$ (see the next paragraph).

For a good plasmonic metal, $Q \gg 1$ (e.g. $Q \sim 10$ – 100 for silver in the entire optical region). For most metal-containing metamaterials, the metal fill factor is not small, typically $f \gtrsim 0.5$. Thus, keeping equation (3.4) in mind, it is very realistic to assume the following condition:

$$\frac{fQ}{s_n(1 - s_n)} \gg 1. \quad (3.10)$$

If so, the second (local) term of the field (3.7) dominates and, with a good precision, the local field is the eigenmode's field,

$$\mathbf{e}(\mathbf{r}, \omega) = i \frac{a_n}{\text{Im } s(\omega_n)} \mathbf{E}_n(\mathbf{r}). \quad (3.11)$$

Substituting equation (3.11) into equation (2.5), we obtain a homogenization formula

$$\bar{\varepsilon}(\omega) = b_n \int_V \varepsilon(\mathbf{r}, \omega) [\mathbf{E}_n(\mathbf{r})]^2 dV, \quad (3.12)$$

where $b_n > 0$ is a real positive coefficient whose specific value is not needed for the purposes of this paper but is given below for the sake of completeness:

$$b_n = \frac{1}{V} \left(\frac{Q \int_V \theta(\mathbf{r}) \mathbf{E}_n(\mathbf{r}) dV}{s_n(1 - s_n)} \right)^2. \quad (3.13)$$

Using equations (3.12) and (3.2) and (3.3), it is straightforward to show that the effective permittivity (3.12) simplifies exactly to

$$\bar{\varepsilon}(\omega) = b_n [s_n \varepsilon_m(\omega) + (1 - s_n) \varepsilon_h(\omega)]. \quad (3.14)$$

4. Conditions of loss compensation by gain and spasing

In the case of the full inversion (maximum gain) and in the exact resonance, the host medium permittivity acquires the imaginary part describing the stimulated emission as given by the standard expression

$$\varepsilon_h(\omega) = \varepsilon_d - i \frac{4\pi |\mathbf{d}_{12}|^2 n_c}{3 \hbar \Gamma_{12}}, \quad (4.1)$$

where $\varepsilon_d = \text{Re } \varepsilon_h$, \mathbf{d}_{12} is a dipole matrix element of the gain transition in a chromophore centre of the gain medium, Γ_{12} is a spectral width of this transition and n_c is the concentration of these centres.¹

The condition for the full electric loss compensation in the metamaterial and amplification (overcompensation) at the resonant frequency $\omega = \omega_n$ is

$$\text{Im } \bar{\varepsilon}(\omega) \leq 0. \quad (4.2)$$

Taking equation (3.14) into account, this reduces to

$$s_n \text{Im } \varepsilon_m(\omega) - \frac{4\pi |\mathbf{d}_{12}|^2 n_c (1 - s_n)}{3 \hbar \Gamma_{12}} \leq 0. \quad (4.3)$$

Finally, taking into account equations (3.4) and (3.5) and that $\text{Im } \varepsilon_m(\omega) > 0$, we obtain from equation (4.3) the condition of the loss (over)compensation as

$$\frac{4\pi |\mathbf{d}_{12}|^2 n_c [1 - \text{Re } s(\omega)]}{3 \hbar \Gamma_{12} \text{Re } s(\omega) \text{Im } \varepsilon_m(\omega)} \geq 1, \quad (4.4)$$

¹If the inversion is not maximum, then this and subsequent equations are still applicable if one sets as the chromophore concentration n_c the inversion density: $n_c = n_2 - n_1$, where n_2 and n_1 are the concentrations of the chromophore centres of the gain medium in the upper and lower states of the gain transition, respectively.

where the strict inequality corresponds to the overcompensation and net amplification. In equation (4.1), we have assumed non-polarized gain transitions. If these transitions are all polarized along the excitation electric field, the concentration n_c should be multiplied by a factor of 3.

Equation (4.4) is a fundamental condition, which is precise (assuming that the requirement (3.10) is satisfied, which is very realistic for metamaterials) and general. Moreover, it is fully analytical and, actually, very simple. Remarkably, it depends only on the material characteristics and does not contain any geometric properties of the metamaterial system or the local fields. In particular, the hot spots, which are prominent in the local fields of nanostructures [35,38], are completely averaged out owing to the integrations in equations (2.5) and (3.12). We note that this implies that taking into account the gain enhancement by a factor $(\epsilon_h + 2)/3$ owing to the local field effects in Wuestner *et al.* [31] is erroneous.

The condition (4.4) is completely non-relativistic (quasi-static)—it does not contain the speed of light c , which is also characteristic of the spaser. It is useful to express this condition also in terms of the stimulated emission cross section $\sigma_e(\omega)$ (where ω is the central resonance frequency) of a chromophore of the gain medium as

$$\frac{c\sigma_e(\omega)\sqrt{\epsilon_d}n_c[1 - \text{Re } s(\omega)]}{\omega \text{Re } s(\omega) \text{Im } \epsilon_m(\omega)} \geq 1. \quad (4.5)$$

It is of fundamental importance to compare this condition of the full loss (over)compensation with the spasing condition [17]. This criterion of spasing, which we will use in the form of eqn (14) of Stockman [20], is fully applicable for the considered metamaterial. For the zero detuning between the gain medium and the SP eigenmode, this criterion can be exactly expressed as [20]

$$\frac{4\pi}{3} \frac{|\mathbf{d}_{12}|^2 \text{Re } s(\omega)}{\hbar \gamma_n \Gamma_{12} \text{Re } s'(\omega)} \int_V |\mathbf{E}_n(\mathbf{r})|^2 \rho(\mathbf{r}) \, dV \geq 1, \quad (4.6)$$

where $\gamma_n = \text{Im } s(\omega)/\text{Re } s'(\omega)$ is the decay rate [17] of the SPs at a frequency ω , $s'(\omega) \equiv \partial s(\omega)/\partial \omega$, and $\rho(\mathbf{r})$ is the density of the gain medium chromophores.

The field quantization in general and SP field quantization in particular can only be carried out consistently when the energy loss is small enough [17]. In our case, this implies that the quality factor is very large, $Q \gg 1$. Otherwise, the field energy needed for the quantization is not conserved and, actually, cannot be introduced [39]. For $Q \gg 1$, we have, with a good accuracy,

$$\gamma_n = \frac{\text{Im } \epsilon_m(\omega)}{\text{Re } \epsilon'_m(\omega)} \quad \text{and} \quad \text{Re } s'(\omega) = \frac{1}{\epsilon_d} [\text{Re } s(\omega)]^2 \text{Re } \epsilon'_m(\omega), \quad (4.7)$$

where $\epsilon'_m(\omega) = \partial \epsilon_m(\omega)/\partial \omega$. Substituting this into equation (4.6), we obtain for the spasing condition

$$\frac{4\pi}{3} \frac{|\mathbf{d}_{12}|^2}{\hbar \Gamma_{12} \text{Re } s(\omega) \text{Im } \epsilon_m(\omega)} \int_V |\mathbf{E}_n(\mathbf{r})|^2 \rho(\mathbf{r}) \, dV \geq 1. \quad (4.8)$$

Taking equations (3.2) and (3.3) into account and assuming that $\rho_n(\mathbf{r}) = [1 - \theta(\mathbf{r})]n_c$, i.e. the chromophores are distributed in the dielectric with a constant density n_c , we *exactly* reduce equation (4.8) to the form of equation (4.4). This brings us to an important conclusion: the full compensation

(overcompensation) of the optical losses in a metamaterial, which is resonant and dense enough to satisfy condition (3.10), and the spasing occur under precisely the same conditions. Inequality (4.4) is then a criterion for both the loss (over)compensation and spasing.

5. Discussion of results and comparison with publications

The above description of a gain metamaterial as a spaser is given for a macroscopic system that is much smaller than the photon wavelength λ . Nevertheless, it is still applicable to any macroscopic metamaterial. The reason is that such a metamaterial is a periodic system that supplies feedback just like the grating is a distributed feedback (DFB) laser. Because it is a plasmonic system, there is no requirement that the periodicity of the grating is a multiple of $\lambda/2$. The lattice constant a of such a DFB spaser can be much smaller than $\lambda/2$. The generating modes of any DFB laser or spaser are band-edge Bloch modes, which are non-propagating and whose half-wavelength is equal to a . Because $a \ll \lambda$, these spasing modes are dark and cannot decay into photons, minimizing their radiative loss. These non-propagating band-edge modes are also known to localize and will spase inside the metamaterial, clamping the inversion and eliminating the net gain. The fact of the equivalence of the full loss compensation and spasing is intimately related to the general criteria of the thermodynamic stability with respect to small fluctuations of electric and magnetic fields (see ch. IX of [39])

$$\text{Im } \bar{\epsilon}(\omega) > 0, \quad \text{Im } \bar{\mu}(\omega) > 0, \quad (5.1)$$

which must be *strict* inequalities for all frequencies for electromagnetically stable systems, i.e. systems stable with respect to small perturbations of the electromagnetic fields. For systems in thermodynamic equilibrium, these conditions are automatically satisfied.

However, for the systems with gain, the conditions (5.1) can be violated, which means that such systems can be electromagnetically unstable. The first of conditions (5.1) is opposite to equations (4.2) and (4.4). This has a transparent meaning: the electrical instability of the system is resolved by its spasing.

The significance of these stability conditions for gain systems can be elucidated by the following *gedanken* experiment. Take a small isolated piece of such a metamaterial. Consider that it is excited at an optical frequency ω either by a weak external optical field \mathbf{E} or acquires such a field owing to fluctuations (thermal or quantum). The energy density² \mathcal{E} of such a system is given by the Brillouin formula [39]

$$\mathcal{E} = \frac{1}{16\pi} \frac{\partial \omega \text{Re } \bar{\epsilon}}{\partial \omega} |\mathbf{E}|^2. \quad (5.2)$$

The internal optical energy density loss Q per unit time (i.e. the rate of the heat density production in the system) is [39]

$$Q = \frac{1}{8\pi} \omega \text{Im } \bar{\epsilon} |\mathbf{E}|^2. \quad (5.3)$$

²For the energy of the system to be definite, it is necessary to assume that the loss is not too large, $|\text{Re } \bar{\epsilon}| \gg \text{Im } \bar{\epsilon}$. This condition is well realistic for most metamaterials.

Assume that the internal (ohmic) loss dominates over other loss mechanisms such as the radiative loss or energy flux out of the selected subsystems owing to propagating modes, which is also a realistic assumption since the ohmic loss is very large for the experimentally studied systems.

In this case of dominating ohmic losses, we have $d\mathcal{E}/dt = Q$. Then equations (5.2) and (5.3) can be resolved together yielding the energy \mathcal{E} and electric field $|\mathbf{E}|$ of this system to evolve with time t exponentially with some decrement Γ as

$$|\mathbf{E}| \propto \sqrt{\mathcal{E}} \propto e^{-\Gamma t} \quad \text{and} \quad \Gamma = \frac{\omega \text{Im} \bar{\epsilon}}{\partial(\omega \text{Re} \bar{\epsilon})/\partial\omega}. \quad (5.4)$$

We are interested in a resonant case when the metamaterial possesses a resonance at some eigenfrequency $\omega_n \approx \omega$. This is true, in particular, for the case of plasmonic behaviour when $\text{Re} \bar{\epsilon}(\omega) < 0$. Then the dominating contribution to $\bar{\epsilon}$ comes from a resonant SP eigenmode n with a frequency $\omega_n \approx \omega$. In such a case, the dielectric function [35] $\bar{\epsilon}(\omega)$ has a simple pole at $\omega = \omega_n$. As a result, $\partial\omega \text{Re} \bar{\epsilon}/\partial\omega \approx \omega \partial \text{Re} \bar{\epsilon}/\partial\omega$ and, consequently, $\Gamma = \gamma_n$, where γ_n is given by equation (4.7), where the metal dielectric function ϵ_m is replaced by the effective permittivity $\bar{\epsilon}$ of the metamaterial. Thus, equation (5.4) is fully consistent with the spectral theory of SPs whose result is equation (4.7).

If the losses are not very large so that energy of the system is meaningful, the Kramers–Kronig causality requires [39] that $\partial(\omega \text{Re} \bar{\epsilon})/\partial\omega > 0$. Thus, $\text{Im} \bar{\epsilon} < 0$ in equation (5.4) would lead to a negative decrement, $\Gamma < 0$, implying that the initial small fluctuation starts exponentially growing in time in its field and energy, which is an instability. Such an instability is indeed not impossible: it will result in spasing that will eventually stabilize $|\mathbf{E}|$ and \mathcal{E} at finite stationary levels of the spaser generation.

Note that the spasing limits (clamps) the gain and population inversion making *the net gain to be precisely zero* [20] in the stationary (continuous wave or CW) regime. This makes the complete loss compensation and its overcompensation impossible in a dense resonant metamaterial where the feedback is created by the internal inhomogeneities and the facets of the system.

To illustrate this point, we show in figure 1 the kinetics of the stationary (CW) operation of a nanoshell spaser [20]. Figure 1*a* displays the SP population N_n per spasing mode as a function of the pumping rate g that shows a pronounced threshold and a linear increase with g typical for both lasers and spasers [20]. This quasi-linearity is due to the fact that the stimulated emission dominates, and the excitation generated in the gain medium with a high probability transfers the energy to the coherent SP population.

The pronounced clamping of the population inversion after the onset of the spasing is illustrated in figure 1*b*. This displays the population inversion $n_{21} = n_2 - n_1$, where n_2 is the population of the upper spasing level in the gain medium, and n_1 is that of the lower level. As one can see from comparison with figure 1*a*, above the threshold of the spasing, the population inversion of the gain medium is clamped at a rather low level $n_{21} \sim 1\%$. The corresponding net amplification in the CW spasing regime is exactly zero, which is a condition for the CW regime.

Because the loss (over)compensation condition (4.4), which is also the spasing condition, is geometry-independent, it is useful to illustrate it for commonly used plasmonic metals, gold and silver whose permittivity we adopt from

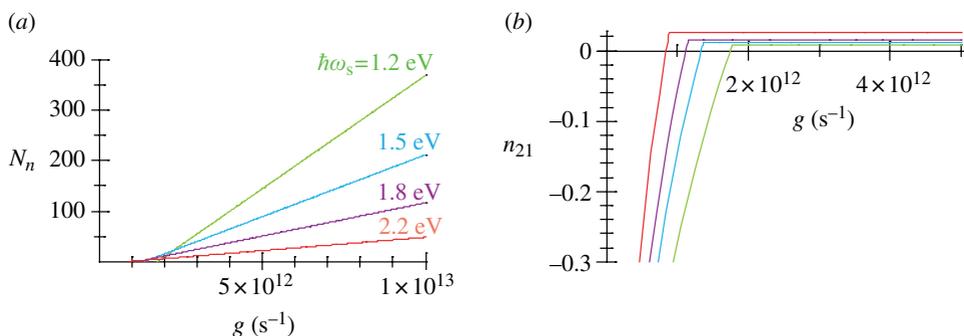


Figure 1. Illustration of the stationary (CW) mode of a nanoshell spaser (adapted from [20]). The curves are colour-coded corresponding to the spasing frequency ω_s determined by the aspect ratio of the corresponding nanoshell. (a) Population of SPs per spasing mode as a function of excitation rate g per one chromophore of the gain medium. (b) Population inversion n_{21} as a function of excitation rate g .

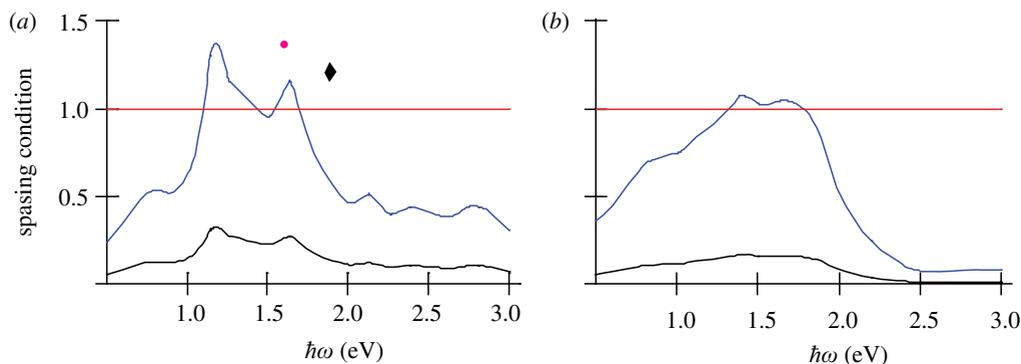


Figure 2. Spasing criterion as a function of optical frequency ω . The straight line (red) represents the threshold for the spasing and full loss compensation, which take place for the curve segments above it. (a) Computations for silver. The chromophore concentration is $n_c = 6 \times 10^{18} \text{ cm}^{-3}$ for the lower curve (black) and $n_c = 2.9 \times 10^{19} \text{ cm}^{-3}$ for the upper curve (blue). The black diamond shows the value of the spasing criterion for the conditions of Noginov *et al.* [40]—see the text. (b) Computations for gold. The chromophore concentration is $n_c = 3 \times 10^{19} \text{ cm}^{-3}$ for the lower curve (black) and $n_c = 2 \times 10^{20} \text{ cm}^{-3}$ for the upper curve (blue).

Johnson & Christy [41]. For the gain-medium chromophores, we will use a reasonable set of parameters, which we will, for the sake of comparison, adapt from Wuestner *et al.* [31]: $\Gamma_{12} = 5 \times 10^{13} \text{ s}^{-1}$ and $d_{12} = 4.3 \times 10^{-18} \text{ esu}$. The results of computations are shown in figure 2. For silver as a metal and $n_c = 6 \times 10^{18} \text{ cm}^{-3}$, the corresponding lower (black) curve in figure 2a does not reach the value of 1, implying that no full loss compensation is achieved. In contrast, for a higher but still very realistic concentration of $n_c = 2.9 \times 10^{19} \text{ cm}^{-3}$, the upper curve in figure 2a does cross the threshold line in the near-infrared region. Above the threshold area, there will be instability and the onset of spasing. As figure 2b demonstrates, for gold the spasing occurs at higher, but still realistic, chromophore concentrations.

Now let us discuss the implications of our results for the research published recently on gain metamaterials. To carry out a quantitative comparison with Wuestner *et al.* [31], we turn to figure 2*a* where the lower (black) curve corresponds to the nominal value of $n_c = 6 \times 10^{18} \text{ cm}^{-3}$ used in Wuestner *et al.* [31]. There is no full loss compensation and spasing. This is explained by the fact that Wuestner *et al.* [31] uses, as a close inspection shows, the gain dipoles parallel to the field (this is equivalent to increasing n_c by a factor of 3) and the local field enhancement (this is equivalent to increasing n_c by a factor of $(\epsilon_h + 2)/3$, which, in actuality, is eliminated owing the space integration—see our discussion in the paragraph after equation (4.4)). This is equivalent to increasing in our formulas the concentration n_c of the chromophores by a factor of $\epsilon_h + 2$ to $n_c = 2.9 \times 10^{19} \text{ cm}^{-3}$, which corresponds to the upper curve in figure 2*a*. This curve rises above the threshold line exactly in the same (infra)red region as in Wuestner *et al.* [31]. Note that this article of Wuestner *et al.* replaces the actual nanolocalized plasmonic field with the spatially uniform Lorentz field $\mathbf{E}_{\text{loc}} = (1/3)(\epsilon_h + 2)\mathbf{E}$, where \mathbf{E} is the macroscopic (external) field that is constant in space. This leads to the loss of any spasing whose feedback is supplied by the spatial variation and nanolocalization of the plasmonic modes.

This agreement of the threshold frequencies between our analytical theory and numerical theory [31] is not accidental: inside the region of stability (i.e. in the absence of spasing) both theories should and do give identical results, provided that the gain-medium transition alignment is taken into account, and the local field-enhancement-effect elimination by the averaging is taken into account. However, above the threshold (in the region of the overcompensation), there should be spasing causing the population inversion clamping and zero net gain, and not a loss compensation. To describe this effect, one has to invoke the equation for coherent SP amplitude (eqn (6) of [20]), which is absent in Wuestner *et al.* [31].

The complete loss compensation is stated in a recent experimental paper [30] where the system was actually a nanofilm rather than a three-dimensional metamaterial. For the rhodamine 800 dye used with extinction cross section [42] $\sigma = 2 \times 10^{-16} \text{ cm}^2$ at 690 nm in concentration $n_c = 1.2 \times 10^{19} \text{ cm}^{-3}$, realistically assuming $\epsilon_d = 2.3$, for frequency $\hbar\omega = 1.7 \text{ eV}$, we calculate from equation (4.5) a point shown by the magenta solid circle in figure 2*a*, which is significantly above the threshold. Because in such a nanostructure the local fields are very non-uniform and confined near the metal similar to the spaser, they likewise cause a feedback. The condition of equation (3.10) is likely to be well satisfied for Xiao *et al.* [30]. Thus, the system should spase, which will cause the clamping of inversion and loss of gain.

In contrast to these theoretical arguments, there is no evidence of spasing indicated in the experiment [30], which can be explained by various factors. Among them, the system of Xiao *et al.* [30] is a gain-plasmonic nanofilm and not a true three-dimensional material. This system is not isotropic. Also, the size of the unit cell of approximately 250 nm is significantly greater than the reduced wavelength λ , which violates the quasi-static conditions and makes the possibility of homogenization and considering this system as an optical metamaterial problematic. This circumstance may also lead to an appreciable spatial dispersion.

In an experimental study of the lasing spaser [26], a nanofilm of PbS QDs was positioned over a two-dimensional metamaterial consisting of an array of negative split-ring resonators. When the QDs were optically pumped, the system exhibited an increase of the transmitted light intensity on the background of a strong luminescence of the QDs but apparently did not reach the lasing threshold. The polarization-dependent loss compensation was only approximately 1 per cent. Similarly, for an array of split-ring resonators over a resonant quantum well, where the inverted electron–hole population was excited optically [29], the loss compensation did not exceed approximately 8 per cent. The relatively low loss compensation in these papers may be due either to random spasing and/or spontaneous or amplified spontaneous emission enhanced by this plasmonic array, which reduces the population inversion.

A dramatic example of possible random spasing is presented in Noginov *et al.* [40]. The system studied was a Kretschmann-geometry SPP set-up [43] with an added approximately 1 μm polymer film containing rhodamine 6G dye in $n_c = 1.2 \times 10^{19} \text{ cm}^{-3}$ concentration. When the dye was pumped, there was outcoupling of radiation in a range of angles. This was a threshold phenomenon with the threshold increasing with the Kretschmann angle. At the maximum of the pumping intensity, the widest range of the outcoupling angles was observed, and the frequency spectrum at every angle narrowed to a peak near a single frequency $\hbar\omega \approx 2.1 \text{ eV}$.

These observations of Noginov *et al.* [40] can be explained by the spasing where the feedback is provided by roughness of the metal. At high pumping, the localized SPs (hot spots), which possess the highest threshold, start to spase in a narrow frequency range around the maximum of the spasing criterion (4.4). Because of the sub-wavelength size of these hot spots, the Kretschmann phase-matching condition is relaxed, and the radiation is outcoupled into a wide range of angles.

The SPPs of Noginov *et al.* [40] excited by the Kretschmann coupling are short-range SPPs, very close to the antisymmetric SPPs. They are localized at sub-wavelength distances from the surface, and their wavelength in the plane is much shorter than ω/c . Thus, they can be well described by the quasi-static approximation and the present theory is applicable to them. Substituting the above-given parameters of the dye and the extinction cross section $\sigma_e = 4 \times 10^{-16} \text{ cm}^2$ into equation (4.5), we obtain a point shown by the black diamond in figure 2*a*, which is clearly above the threshold, supporting our assertion of the spasing. Likewise, the amplified spontaneous emission and, possibly, spasing appear to have prevented the full loss compensation in a SPP system of Bolger *et al.* [28].

Note that the long-range SPPs of Leon & Berini [27] are localized significantly weaker (at distances approx. λ) than those excited in Kretschmann geometry. Thus, the long-range SPPs experience a much weaker feedback, and the amplification instead of the spasing can be achieved. Generally, the long-range SPPs are fully electromagnetic (non-quasi-static) and are not describable in the present theory.

Concluding, we have fundamentally established that the conditions of the full loss compensation (overcompensation) and spasing in dense resonant plasmonic metamaterials (satisfying the realistic condition of equation (3.10)) are identical. These conditions are analytical and universal, i.e. independent from the metamaterial geometry. Owing to the feedback inherent in the dense

resonant metamaterials, this implies that an attempt at the full loss compensation (over-compensation) in such metamaterials in actuality causes spasing that clamps the gain-medium population inversion and eliminates the net gain, precluding the full loss compensation.

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