Spaser Action, Loss Compensation, and Stability in Plasmonic Systems with Gain

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We demonstrate that the conditions of spaser generation and the full loss compensation in a resonant plasmonic-gain medium (metamaterial) are identical. Consequently, attempting the full compensation or overcompensation of losses by gain will lead to instability and a transition to a spaser state. This will limit (clamp) the inversion and lead to the limitation on the maximum loss compensation achievable. The criterion of the loss overcompensation, leading to the instability and spasing, is given in an analytical and universal (independent from system’s geometry) form.

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There is a tremendous interest in nanoplasmonic systems with gain, which has been initiated by the introduction of the spaser\(^1\). Such systems consist of a metal nanoplasmonic component and a nanoscale gain medium (dye molecules, quantum dots, etc.) where the population inversion is created optically or electrically\(^1\). If the surface plasmon (SP) amplification by stimulated emission overcomes the loss, the initial state of the system loses its stability, and a new, spasing state appears due to a spontaneous symmetry breaking\(^3\). The spaser is a nanoscopic generator of coherent local optical fields and their ultrafast nanoamplifier.

There has been an active development of the idea of spaser. A nanoscopic spaser consisting of gold nanosphere surrounded by a dielectric gain shell containing a laser dye has been demonstrated\(^1\). Surface plasmon polariton (SPP) spasers have been demonstrated with one-, two-, and three-dimensional confinement.\(^5\) The pre-generation narrowing of the resonant line in the lasing spaser has been observed\(^8\).

One of the most active research directions related to the spaser has been compensation of losses by gain in plasmonic waveguides and metamaterials, which is of principal importance due to high losses in the optical range of frequencies. Amplification of long-range SPPs in a gold strip waveguide in the proximity of a pumped dye solution has been demonstrated\(^9\). Amplified spontaneous emission of SPPs has been observed in gold nanofilms over an amplifying medium containing PbS quantum dots, where the reduction of the SPP propagation loss by up to 30 percent has taken place\(^10\). In a metamaterial consisting of split-ring resonators coupled to an optically-pumped InGaAs quantum well, a reduction of the transmission loss by \(\approx 8\) percent has been observed\(^11\). The full compensation and overcompensation of the optical transmission loss for a fishnet metamaterial containing a pumped dye dispersed in a polymer matrix has been observed\(^12\). This experiments has later been stated to be in agreement with a theory based on a Maxwell-Bloch equations\(^13\).

In this Letter we show that the full compensation or overcompensation of the optical loss in an active metamaterial (i.e., a nanostructured optical system of a finite size containing a gain medium) leads to an instability that is resolved by its spasing (i.e., due to becoming a spaser). We further show that the conditions of the complete resonant gain compensation (which is the only one explored either experimentally or theoretically so far) and the threshold condition of spasing are identical. This spasing limits (clamps) the gain and, consequently, does not allow for the complete loss compensation (overcompensation) at any frequency. Additionally, this spasing in the gain metamaterial will show itself as enhanced (amplified) spontaneous emission and the coherent emission reminding the lasing spaser\(^8,14\).

We will consider, for certainty, an isotropic and uniform metamaterial that, by definition, in a range of frequencies \(\omega\) can be described by the effective permittivity \(\varepsilon(\omega)\) and permeability \(\mu(\omega)\). 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 sizes much greater than the unit cell but much smaller than the wavelength \(\lambda\), which is a metamaterial itself. Let us subject this metamaterial to a uniform electric field \(E(\omega)\) oscillating with frequency \(\omega\). We will denote the local field at a point \(r\) inside this metamaterial as \(e(r,\omega)\). For such a small piece of the metamaterial, a homogenization procedure gives an exact expression (see Ref. 15 and references cited therein)

\[
\varepsilon(\omega) = \frac{1}{V |E(\omega)|^2} \int_V \varepsilon(r,\omega) |e(r,\omega)|^2 d^3r ,
\]

where \(V\) is the volume of the metamaterial piece.

Consider a frequency \(\omega\) close to the resonance frequency \(\omega_n\) of an nth plasmonic eigenmode. To be bright, this eigenmode must be dipolar. Then the Green’s function expansion\(^16,17\) shows that the eigenmode’s field can be estimated as \(\sim EQf\), where \(f\) is the fill factor of the metal component, and \(Q = -\text{Re} \varepsilon_m(\omega)/\text{Im} \varepsilon_m(\omega)\) with the metal’s permittivity \(\varepsilon_m(\omega)\). Realistically assuming that \(fQ \gg 1\), we conclude that the resonant eigenmode’s field \(E_n(r) = -\nabla \varphi_n(r)\) dominates the local field, \(e(r,\omega) \approx a_n E_n(r)\), where \(a_n\) is a constant whose
exact value we will not need. In this case, the effective permittivity (1) becomes
\[
\varepsilon(\omega) = |a_n|^2 \int_V \varepsilon(\mathbf{r}, \omega) |\mathbf{E}_n(\mathbf{r})|^2 \, d^3 r .
\] (2)

Note that we conventionally assume an eigenmode normalization: \( \int_V |\mathbf{E}_n(\mathbf{r})|^2 \, d^3 r = 1. \)

The quasistatic eigenmode equation is\(^{18}\)
\[
\nabla \theta(\mathbf{r}) \nabla \varphi_n(\mathbf{r}) = s_n \nabla^2 \varphi_n(\mathbf{r}) ,
\] (3)
where \( s_n \) 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 Eq. (3) one can easily find that
\[
s_n = \int_V \theta(\mathbf{r}) |\mathbf{E}_n(\mathbf{r})|^2 \, d^3 r , \quad 1 \geq s_n \geq 0 . \quad (4)
\]
The resonant frequency, \( \omega = \omega_n \), is defined by
\[
s_n = \text{Re} \, s(\omega) , \quad s(\omega) = \frac{\varepsilon_h(\omega)}{\varepsilon_h(\omega) - \varepsilon_m(\omega)} , \quad (5)
\]
where \( s(\omega) \) is Bergman’s spectral parameter, and \( \varepsilon_h(\omega) \) is permittivity of the surrounding host containing the gain chromophore centers.

In the case of the full inversion (maximum gain) and in the exact resonance, the host medium permittivity acquires the imaginary part responsible for the stimulated emission as given by the standard expression
\[
\varepsilon_h(\omega) = \varepsilon_d - \frac{4\pi |\mathbf{d}_{12}|^2 n_c}{3 \hbar \Gamma_{12}} .
\] (6)

where \( \varepsilon_d = \text{Re} \varepsilon_h \), \( \mathbf{d}_{12} \) is a dipole matrix element of the gain transition in a chromophore center of the gain medium, \( \Gamma_{12} \) is a spectral width of this transition, and \( n_c \) is the concentration of these centers.

Using Eqs. (2) and (4), it is straightforward to show that the effective permittivity (2) simplifies exactly to
\[
\varepsilon(\omega) = |a_n|^2 [s_n \varepsilon_m(\omega) + (1-s_n)\varepsilon_h(\omega)] .
\] (7)

The condition for the full electric loss (over)compensation at the resonant frequency \( \omega = \omega_n \) is \( \text{Im} \, \varepsilon(\omega) \leq 0 \), which reduces to
\[
s_n \text{Im} \varepsilon_m(\omega) - \frac{4\pi |\mathbf{d}_{12}|^2 n_c (1-s_n)}{3 \hbar \Gamma_{12}} \leq 0 .
\] (8)

Finally, taking into account Eqs. (4)-(5) and that \( \text{Im} \varepsilon_m(\omega) > 0 \), we obtain from Eq. (8) the condition of the loss (over)compensation as
\[
\frac{4\pi |\mathbf{d}_{12}|^2 n_c |1 - \text{Re} s(\omega)|}{3 \hbar \Gamma_{12} \text{Re} s(\omega) \text{Im} \varepsilon_m(\omega)} \geq 1 ,
\] (9)
where the strict inequality corresponds to the overcompensation and net amplification. In Eq. (6) 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.

This is a fundamental condition, which is precise (for \( Qf \gg 1 \)) and general. 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\(^{18,19}\), are completely averaged out due to the integrations in Eqs. (1) and (2). This implies that taking into account the gain enhancement due to the local field effects in Ref. 13 is erroneous.

The condition (9) is completely non-relativistic (quasistatic) – it does not contain speed of light \( c \), which is characteristic of the spaser. It is useful to express this condition also in terms of the total extinction cross section \( \sigma(\omega) \) (where \( \omega \) is the central resonance frequency) of a chromophore of the gain medium as
\[
\frac{\sigma(\omega) \sqrt{\varepsilon_m n_c} [1 - \text{Re} s(\omega)]}{\omega \text{Re} s(\omega) \text{Im} \varepsilon_m(\omega)} \geq 1 .
\] (10)

It is of fundamental importance to compare this condition of the full loss (over)compensation with the spasing condition\(^1\). This criterion of spasing, which we will use in the form of Eq. (14) of Ref. 3, 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\(^3\)
\[
\frac{4\pi |\mathbf{d}_{12}|^2 \text{Re} s(\omega)}{\hbar \gamma_n \Gamma_{12} \text{Re} s'(\omega)} \int_V |\mathbf{E}_n(\mathbf{r})|^2 \rho(\mathbf{r}) d^3 r \geq 1 .
\] (11)

where \( \gamma_n = \text{Im} s(\omega) / \text{Re} s'(\omega) \) is the decay rate\(^1\) of the SPs at a frequency \( \omega \), \( s'(\omega) \equiv \partial s(\omega) / \partial \omega \), and \( \rho(\mathbf{r}) \) is the density of the gain medium chromophores.

The SP field quantization can only be carried out consistently when the energy loss is small enough\(^1\). This implies that the quality factor \( Q \gg 1 \). Otherwise the field energy needed for the quantization is not conserved and, actually, cannot be introduced\(^20\). For \( Q \gg 1 \), we have, with a good accuracy,
\[
\gamma_n = \frac{\text{Im} \varepsilon_m(\omega)}{\text{Re} \varepsilon'_m(\omega)} , \quad \text{Re} s'(\omega) = \frac{1}{\varepsilon_d} [\text{Re} s(\omega)]^2 \text{Re} \varepsilon'_m(\omega) ,
\] (12)
where \( \varepsilon'_m(\omega) = \partial \varepsilon_m(\omega) / \partial \omega \). Substituting this into Eq. (11), we obtain for the spasing condition
\[
\frac{4\pi}{3} \frac{|\mathbf{d}_{12}|^2}{\hbar \Gamma_{12} \text{Re} s(\omega) \text{Im} \varepsilon_m(\omega)} \int_V |\mathbf{E}_n(\mathbf{r})|^2 \rho(\mathbf{r}) d^3 r \geq 1 .
\] (13)

Taking Eq. (4) 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
Eq. (13) to the form of Eq. (9). This brings us to an important conclusion: the full compensation (overcompensation) of the optical losses in a resonant dense metamaterial with $fQ \gg 1$ and the spasing occur under precisely the same conditions. Inequality (9) is the criterion for both the loss (over)compensation and spasing.

This 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 Chap. IX of Ref. 20)

$$\text{Im} \bar{\varepsilon}(\omega) > 0 , \quad \text{Im} \bar{\mu}(\omega) > 0 , \quad (14)$$

which must be strict inequalities for all frequencies.

The first of these conditions is opposite to Eq. (9). This has a transparent meaning: the electrical instability of the system is resolved by its spasing that limits (clamps) the gain and population inversion making the net gain to be precisely zero$^3$. This makes the complete loss compensation and its overcompensation impossible in a metamaterial with a feedback, which is created by the facets of the system and its internal inhomogeneities.

Because the loss (over)compensation condition (9), which is also the spasing condition, is geometry-independent, it is useful to illustrate it for commonly used plasmonic metals, gold and silver$^{21}$. For the gain medium chromophores, we will use a reasonable set of parameters, which we will, for the sake of comparison, adapt from Ref. 13: $F_{12} = 5 \times 10^{13}$ s$^{-1}$ and $d_{12} = 4 \times 10^{-18}$ esu. The results of computations are shown in Fig. 1. For silver as a metal and $n_e = 6 \times 10^{18}$ cm$^{-3}$, the corresponding lower (black) curve in panel (a) 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 $n_e = 3.9 \times 10^{19}$ cm$^{-3}$, the upper curve in Fig. 1 (a) does cross the threshold line in the near-infrared region. Above the threshold area, there will be the instability and the onset of the spasing. As Fig. 1 (b) demonstrates, for gold the spasing is at higher, but still realistic, chromophore concentrations.

Now let us discuss the implications of our results for the research published recently on the gain metamaterials. We start with the recent theoretical paper$^{13}$ that summarizes "... We show that appropriate placing of optically pumped laser dyes (gain) into the metamaterial structure results in a frequency band where the nonbimolecular gain becomes amplifying. In that region both the real and the imaginary part of the effective refractive index become simultaneously negative and the figure of merit diverges at two distinct frequency points."$^3$

In light of the present results, this statement is incorrect. In reality, such a regime in Ref. 13 brings about the instability resulting in spasing for the region of negative loss, $\text{Im} \bar{\varepsilon} \leq 0$, which is interpreted in Ref. 13 as the loss compensation. The full quantum mechanical theory$^3$ of the spasing in gain nanoplasmic systems shows that within the band of the spasing or (over)compensation, the population inversion is not defined by pumping and cannot be arbitrarily large. It is determined self-consistently by the processes of the simulated emission of SPs and their relaxation. The stationary spasing decreases the inversion until it eliminates the net gain completely$^3$. Theory of Ref. 13 misses an equation for the coherent SP field and, therefore, fails to describe the onset of spasing, which is a non-equilibrium second-order phase transition.

To carry out a quantitative comparison with Ref. 13, we turn to Fig. 1 (a) where the lower (black) curve corresponds to the nominal value of $n_e = 6 \times 10^{18}$ cm$^{-3}$ used in Ref. 13. There is no full loss compensation and spasing, which is explained by the fact that Ref. 13 uses, as a close inspection shows, the gain dipoles parallel to the field and the local field enhancement [the latter, actually, is eliminated by the space integration – see our discussion after Eq. (9)]. This is equivalent to increasing in our formulas the concentration of the chromophores to $n_e = 3.9 \times 10^{19}$ cm$^{-3}$, which corresponds to the upper curve in Fig. 1 (a). This curve rises above the threshold line exactly in the same (infra)red region as in Ref. 13. In reality, above the threshold there will be spasing causing the zero net gain and not a loss compensation.

The complete loss compensation is claimed in the re-
cent experimental paper\(^{12}\) where the system was actually a nanofilm rather than a three-dimensional metamaterial. The effective index parameters have been found from the transmission data by comparison to theory. For the Rhodamine 800 dye used with extinction cross section \(\sigma = 2 \times 10^{-16} \text{cm}^2\) at 690 nm\(^{23}\) in concentration \(n_c = 1.2 \times 10^{19} \text{cm}^{-3}\), realistically assuming \(\varepsilon_r = 2.3\), for frequency \(\hbar \omega = 1.7 \text{eV}\) we calculate from Eq. (10) a point shown by the magenta solid circle in Fig. 1 (a), which is significantly above the threshold. Because in such a nanostructure the local fields are very non-uniform and confined near the metal like in the spaser, they likewise cause a feedback. Thus, the system should spase, which will cause the the clamping of inversion and loss of gain.

A dramatic example of possible random spasing is presented in Ref. 22. The system studied was a Kretschmann-geometry setup\(^{24}\) with an added \(\sim 1 \mu\text{m}\) polymer film containing Rhodamine 6G dye in the \(n_c = 1.2 \times 10^{19} \text{cm}^{-3}\) 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 pumped, there was outcoupling of radiation in a range of angles. This was a threshold phenomenon 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 \(\hbar \omega \approx 2.1 \text{eV}\). This can be explained by the spasing where the feedback is provided by roughness of the metal. At the high pumping, the localized SPs with the highest threshold start to spase near a single frequency. Because of their sub-wavelength size, the Kretschmann phase-matching condition is relaxed, and the radiation is outcoupled into a wide range of angles. Substituting the above-given parameters of the dye and \(\sigma = 4 \times 10^{-16} \text{cm}^2\) into Eq. (10), we obtain a point shown by the black diamond in Fig. 1, 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 Ref. 10.

Concluding, we have fundamentally established that the conditions of the full loss compensation (overcompensation) and spasing in dense, resonant plasmonic metamaterials are identical. This condition is analytical and universal, i.e., independent from the metamaterial geometry. Due to the feedback inherent in the inhomogeneous metamaterials and waveguides, this implies that an attempt of the full loss compensation (over-compensation) in actuality brings about spasing that eliminates the net gain and precludes the full loss compensation.

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