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 dense 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 initiated by the introduction of the spaser [1]. Such systems consist of a metal nanoplasmonic component and a nanoscale gain medium (dye molecules, semiconductor nanostructures such as quantum dots, etc.) where the population inversion is created optically or electrically [1,2]. 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 with a coherent SP population whose phase is due to 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 a spaser. A nanoscopic spaser consisting of a gold nanosphere surrounded by a dielectric gain shell containing a laser dye has been demonstrated [4]. Surface plasmon polariton (SPP) spasers have been demonstrated with one-, two-, and three-dimensional confinement [5–7]. The pregeneration 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]. In gold nanofilms over an amplifying medium containing PbS quantum dots, the reduction of the SPP propagation loss by up to 30% has been found [10]. In a metamaterial consisting of split-ring resonators coupled to an optically pumped InGaAs quantum well, a reduction of the transmission loss by ≈ 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]. Furthermore, it has been claimed that this experiment is 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., an optical system of macroscopic dimensions containing a dense metal nanostructure and a gain medium) leads to an instability that is resolved by its spasing (i.e., becoming a spaser). This theory is based on the density matrix equations, which are equivalent to the Maxwell-Bloch equations of Ref. [13], supplemented by an equation for a coherent SP field [3] and Green's function approach [14,15]. We further show that the conditions of the complete resonant gain compensation in the dense nanoplasmonic systems (which is the only type 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 coherent emission similar to the lasing spaser [8,16].

We will consider, for certainty, an isotropic and uniform metamaterial whose unit cell is much smaller than the reduced wavelength $\lambda\!\!\!^{-}$. Then in a range of frequencies ω, it can be described by the effective permittivity $\bar{\varepsilon}(\omega)$ 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 this metamaterial with sizes much greater that the unit cell but much smaller than $\lambda\!\!\!^{-}$, which is a metamaterial itself. Let us subject this metamaterial to a uniform electric field $\mathbf{E}(\omega)$ oscillating with frequency ω. We will denote the local field at a point $\mathbf{r}$ inside this metamaterial as $\mathbf{e}(\mathbf{r}, \omega)$. For such a small piece of the metamaterial, a homogenization procedure gives an exact expression (see Ref. [17] and references cited therein)

$$\bar{\varepsilon}(\omega) = \frac{1}{V|E(\omega)|^2} \int_V \varepsilon(\mathbf{r}, \omega)|\mathbf{e}(\mathbf{r}, \omega)|^2 d^3r, \qquad (1)$$

where V is the volume of this metamaterial piece.

Consider frequency ω close to the resonance frequency ω_n of an nth plasmonic eigenmode. To be bright, this eigenmode must be dipolar. Then the Green's function expansion [14,15] shows that the eigenmode's field can be estimated as $\sim EQf$, where f is the fill factor of the metal component, $Q = -\mathrm{Re}\varepsilon_m(\omega)/\mathrm{Im}\varepsilon_m(\omega)$ is the metal's quality factor, and $\varepsilon_m(\omega)$ is the metal's permittivity. Realistically assuming that $fQ \gg 1$, we conclude that the resonant eigenmode's field $\mathbf{E}_n(\mathbf{r}) = -\nabla\varphi_n(\mathbf{r})$ dominates the local field, $\mathbf{e}(\mathbf{r}, \omega) \approx a_n\mathbf{E}_n(\mathbf{r})$, where a_n is a constant whose exact value we will not need. In this case, the effective permittivity (1) becomes

$$\bar{\varepsilon}(\omega) = |a_n|^2 \int_V \varepsilon(\mathbf{r}, \omega)|\mathbf{E}_n(\mathbf{r})|^2 d^3r. \tag{2}$$

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

The quasistatic eigenmode equation is [18]

$$\nabla\theta(\mathbf{r})\nabla\varphi_n(\mathbf{r}) = s_n\nabla^2\varphi_n(\mathbf{r}), \tag{3}$$

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

$$s_n = \int_V \theta(\mathbf{r})|\mathbf{E}_n(\mathbf{r})|^2 d^3r, \qquad 1 \geq s_n \geq 0. \tag{4}$$

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

$$s_n = \mathrm{Re}s(\omega), \qquad s(\omega) \equiv \frac{\varepsilon_h(\omega)}{\varepsilon_h(\omega) - \varepsilon_m(\omega)}, \tag{5}$$

where $s(\omega)$ is Bergman's spectral parameter, and $\varepsilon_h(\omega)$ is the 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 due to the stimulated emission as given by the standard expression

$$\varepsilon_h(\omega) = \varepsilon_d - i\frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2 n_c}{\hbar\Gamma_{12}}, \tag{6}$$

where $\varepsilon_d = \mathrm{Re}\varepsilon_h$, $\mathbf{d}_{12}$ is the dipole matrix element of the gain transition in a chromophore center of the gain medium, Γ_{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

$$\bar{\varepsilon}(\omega) = |a_n|^2[s_n\varepsilon_m(\omega) + (1 - s_n)\varepsilon_h(\omega)]. \tag{7}$$

The condition for the full electric loss (over)compensation at the resonant frequency $\omega = \omega_n$ is $\mathrm{Im}\bar{\varepsilon}(\omega) \leq 0$, which reduces to

$$s_n\mathrm{Im}\varepsilon_m(\omega) - \frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2 n_c(1 - s_n)}{\hbar\Gamma_{12}} \leq 0. \tag{8}$$

Finally, taking into account Eqs. (4) and (5) and that $\mathrm{Im}\varepsilon_m(\omega) > 0$, we obtain from Eq. (8) the condition of the loss (over)compensation as

$$\frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2 n_c[1 - \mathrm{Re}s(\omega)]}{\hbar\Gamma_{12}\mathrm{Re}s(\omega)\mathrm{Im}\varepsilon_m(\omega)} \geq 1, \tag{9}$$

where the strict inequality corresponds to the overcompensation and net amplification. In Eq. (6) we have assumed nonpolarized 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 nonrelativistic (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 $\sigma_e(\omega)$ (where ω is the central resonance frequency) of a chromophore of the gain medium as

$$\frac{c\sigma(\omega)\sqrt{\varepsilon_d}n_c[1 - \mathrm{Re}s(\omega)]}{\omega\mathrm{Re}s(\omega)\mathrm{Im}\varepsilon_m(\omega)} \geq 1. \tag{10}$$

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]

$$\frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2\mathrm{Re}s(\omega)}{\hbar\gamma_n\Gamma_{12}\mathrm{Re}s'(\omega)}\int_V |\mathbf{E}_n(\mathbf{r})|^2\rho(\mathbf{r})d^3r \geq 1 \tag{11}$$

where $\gamma_n = \mathrm{Im}s(\omega)/\mathrm{Re}s'(\omega)$ is the decay rate [1] 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 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,

$$\gamma_n = \frac{\mathrm{Im}\varepsilon_m(\omega)}{\mathrm{Re}\varepsilon'_m(\omega)}, \qquad \mathrm{Re}s'(\omega) = \frac{1}{\varepsilon_d}[\mathrm{Re}s(\omega)]^2\mathrm{Re}\varepsilon'_m(\omega), \tag{12}$$

where $\varepsilon'_m(\omega) = \partial\varepsilon_m(\omega)/\partial\omega$. Substituting this into Eq. (11), we obtain for the spasing condition

$$\frac{4\pi}{3}\frac{|\mathbf{d}_{12}|^2}{\hbar\Gamma_{12}\mathrm{Re}s(\omega)\mathrm{Im}\varepsilon_m(\omega)}\int_V |\mathbf{E}_n(\mathbf{r})|^2\rho(\mathbf{r})d^3r \geq 1. \quad (13)$$

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])

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

which must be *strict* inequalities for all frequencies in thermodynamic equilibrium.

For gain systems, these conditions may be violated. The first of conditions (14) is opposite to Eq. (9). This has a transparent meaning: when the spasing condition is satisfied, the system possesses the electrical instability, which is resolved by its spasing that limits (clamps) the gain and population inversion turning the net gain to be precisely zero [3]. This makes the complete loss compensation and its overcompensation impossible in a dense resonant metamaterial with a feedback, which is created by the internal inhomogeneities of the system (and its facets too). Precisely the same situation is true for the conventional lasers whose net gain is exactly zero for the stationary generation due to the lasing transition saturation.

Because the loss (over)compensation condition (9), which is also the spasing condition, is geometry independent, it is useful to illustrate it for gold and silver, commonly used plasmonic metals, whose permittivities are adapted from Ref. [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]: $\Gamma_{12} = 5\times10^{13}$ s^{-1} and $d_{12} = 4\times10^{-18}$ esu. The results of computations are shown in Fig. 1. For silver as a metal and $n_c = 6\times10^{18}$ 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 $n_c = 2.9\times10^{19}$ cm^{-3}, the upper curve in Fig. 1(a) does cross the threshold line in the near-infrared region. Above the threshold, there will be the instability and the spasing. As Fig. 1(b) demonstrates, for gold the overcompensation and spasing take place 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. To carry out a quantitative comparison with Ref. [13], we turn to Fig. 1(a) where the lower (black) curve corresponds to

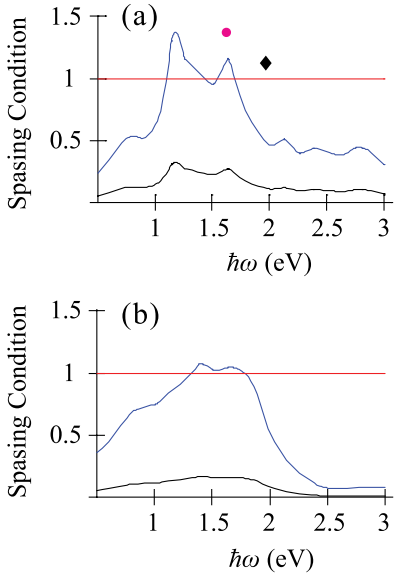

FIG. 1 (color online). Spasing criterion of Eq. (9) as a function of optical frequency ω. The straight red line represents the threshold for the spasing and full loss compensation, which take place for the curve segments above it. (a) Computations for silver. The chromophore concentration is $n_c = 6\times10^{18}$ cm^{-3} for the lower curve (black) and $n_c = 2.9\times10^{19}$cm^{-3} for the upper curve (blue). The magenta solid circle and black diamond show the values of the spasing criterion for the conditions of Refs. [12,23], respectively—see the text. (b) Computations for gold. The chromophore concentration is $n_c = 3\times10^{19}$ cm^{-3} for the lower curve (black) and $n_c = 2\times10^{20}$ cm^{-3} for the upper curve (blue).

the nominal value of $n_c = 6\times10^{18}$ 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 by a factor of $\varepsilon_h + 2$ to $n_c = 2.9\times10^{19}$ 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]. The agreement of the threshold frequencies between our analytical theory and numerical theory [13] 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. This effect is described by the equation for coherent SP amplitude [Eq. (6) of Ref. [3]], which is absent in Ref. [13].

The complete loss compensation is stated in the recent experimental paper [12] where the system was actually a

nanofilm rather than a $3d$ metamaterial. For the Rhodamine 800 dye used with extinction cross section $\sigma = 2 \times 10^{-16}$ cm^2 at 690 nm [22] in concentration $n_c = 1.2 \times 10^{19}$ cm^{-3}, realistically assuming $\varepsilon_d = 2.3$, for frequency $\hbar\omega = 1.7$ 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 nonuniform and confined near the metal like in the spaser, they likewise cause a feedback. Thus, the system should spase, which will cause the clamping of inversion and loss of gain. There is no evidence of spasing indicated in the paper, which can be explained by various factors. Among them, the system of Ref. [12] is a gain-plasmonic nanolayer and not a true $3d$ material. This system is not isotropic. Also, the size of the unit cell $\sim$250 nm is greater than λ, which violates the quasistatic conditions and makes the possibility of homogenization and considering it as an optical metamaterial problematic. This can also lead to an appreciable spatial dispersion.

A dramatic example of possible random spasing is presented in Ref. [23]. The system studied was a Kretschmann-geometry setup [24] with an added ~ 1 μm polymer film containing Rhodamine 6 G dye in the $n_c = 1.2 \times 10^{19}$ 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 $\hbar\omega \approx 2.1$ eV. This can be explained by the spasing where the feedback is provided by roughness of the metal. (The short-range SPPs excited in the Kretschmann geometry are almost quasistatic and can be described by the present theory.) At the high pumping, the localized SPs with the highest threshold start to spase near a single frequency. Because of their subwavelength 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 $\sigma = 4 \times 10^{-16}$ cm^2 into Eq. (10), we obtain a point shown by the black diamond in Fig. 1, which is clearly above the threshold, supporting a possibility of the spasing. It is also possible that spasing prevented the full loss compensation in a SPP system [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. Because of the feedback inherent in the dense resonant metamaterials due to their inhomogeneity on the nanoscale, this implies that an attempt of the full loss compensation (overcompensation) will in actuality bring about spasing thus eliminating the net gain and precluding the full loss compensation.

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[1] D. J. Bergman and M. I. Stockman, Phys. Rev. Lett. **90**, 027402 (2003).
[2] M. I. Stockman and D. J. Bergman, U.S. Patent No. 7 569 188 (2009).
[3] M. I. Stockman, J. Opt. **12**, 024004 (2010).
[4] M. A. Noginov, G. Zhu, A. M. Belgrave, R. Bakker, V. M. Shalaev, E. E. Narimanov, S. Stout, E. Herz, T. Suteewong, and U. Wiesner, Nature (London) **460**, 1110 (2009).
[5] M. T. Hill, M. Marell, E. S. P. Leong, B. Smalbrugge, Y. Zhu, M. Sun, P. J. van Veldhoven, E. J. Geluk, F. Karouta, and Y.-S. Oei *et al.*, Opt. Express **17**, 11 107 (2009).
[6] R. F. Oulton, V. J. Sorger, T. Zentgraf, R.-M. Ma, C. Gladden, L. Dai, G. Bartal, and X. Zhang, Nature (London) **461**, 629 (2009).
[7] R.-M. Ma, R. F. Oulton, V. J. Sorger, G. Bartal, and X. Zhang, Nature (London) **10**, 110 (2010).
[8] E. Plum, V. A. Fedotov, P. Kuo, D. P. Tsai, and N. I. Zheludev, Opt. Express **17**, 8548 (2009).
[9] I. D. Leon and P. Berini, Nat. Photon. **4**, 382 (2010).
[10] P. M. Bolger, W. Dickson, A. V. Krasavin, L. Liebscher, S. G. Hickey, D. V. Skryabin, and A. V. Zayats, Opt. Lett. **35**, 1197 (2010).
[11] N. Meinzer, M. Ruther, S. Linden, C. M. Soukoulis, G. Khitrova, J. Hendrickson, J. D. Olitzky, H. M. Gibbs, and M. Wegener, Opt. Express **18**, 24140 (2010).
[12] S. Xiao, V. P. Drachev, A. V. Kildishev, X. Ni, U. K. Chettiar, H.-K. Yuan, and V. M. Shalaev, Nature (London) **466**, 735 (2010).
[13] S. Wuestner, A. Pusch, K. L. Tsakmakidis, J. M. Hamm, and O. Hess, Phys. Rev. Lett. **105**, 127401 (2010).
[14] M. I. Stockman, S. V. Faleev, and D. J. Bergman, Phys. Rev. Lett. **88**, 067402 (2002).
[15] X. Li and M. I. Stockman, Phys. Rev. B **77**, 195109 (2008).
[16] N. I. Zheludev, S. L. Prosvirnin, N. Papasimakis, and V. A. Fedotov, Nat. Photon. **2**, 351 (2008).
[17] M. I. Stockman, K. B. Kurlayev, and T. F. George, Phys. Rev. B **60**, 17071 (1999).
[18] M. I. Stockman, S. V. Faleev, and D. J. Bergman, Phys. Rev. Lett. **87**, 167401 (2001).
[19] M. I. Stockman, L. N. Pandey, and T. F. George, Phys. Rev. B **53**, 2183 (1996).
[20] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford and New York, 1984).
[21] P. B. Johnson and R. W. Christy, Phys. Rev. B **6**, 4370 (1972).
[22] Z. Gryczynski, O. O. Abugo, and J. R. Lakowicz, Anal. Biochem. **273**, 204 (1999).
[23] M. A. Noginov, G. Zhu, M. Mayy, B. A. Ritzo, N. Noginova, and V. A. Podolskiy, Phys. Rev. Lett. **101**, 226806 (2008).
[24] E. Kretschmann and H. Raether, Z. Naturforsch. A **23**, 2135 (1968).