Amplification of surface plasmon polaritons in the presence of nonlinearity and spectral signatures of threshold crossover

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We describe the effects of nonlinearity on propagation of surface plasmon polaritons (SPPs) at an interface between a metal and an amplifying medium of the externally pumped two-level atoms. Using Maxwell equations we derive the nonlinear dispersion law and demonstrate that the nonlinear saturation of the linear gain leads to formation of stationary SPP modes with the intensities independent from the propagation distance. Transition to the regime of stationary propagation is similar to the threshold crossover in lasers and leads to narrowing of the SPP spectrum. © 2009 Optical Society of America

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Delivery of electromagnetic energy from macro- to nanoscales and its direct generation in nanostructures are the challenging problems of nanophotonics. They are particularly important for plasmonics and metamaterials, which promise subwavelength localization and advanced control of light in photonic nanocircuits. However, metal structures typically suffer from large intrinsic (ohmic) losses hampering attractive applications. On the other hand, the electromagnetic field confinement and enhancement associated with surface plasmons have been proposed to control excitation of active emitters and achieve a plasmonic laser (spaser) [1,2]. Propagating surface plasmon polaritons (SPPs) at an interface with an amplifying dielectric material have been studied theoretically, see, e.g., [3,4] and references therein, and practically demonstrated using optically pumped dyes [5–7] and erbium-doped glass [8]. In particular, a distinct threshold in the dependence of the output SPP intensity on gain and the simultaneous significant narrowing of SPP spectra have been reported inferring signatures of stimulated SPP emission [7].

Theory of SPPs interacting with an amplifying medium has been so far limited to the linear case [1,3,4,6,7]. This approximation describes the SPP modes, where the linear growth of the amplified SPPs is not balanced by the nonlinear losses. In the linear theory plasmons with the real propagation constant (in the SPP case) or the real frequency of the oscillations (in the spaser case) can be found only at the threshold, where the linear loss is exactly balanced by the linear gain [3]. However, in practice, the gain-loss balance should be maintained even above the threshold. This is because the amplification of SPPs is expected to be fully compensated by the nonlinear losses ensuring existence of SPPs with the real propagation constants.

Our main aim here is to present a theory of SPP amplification accounting for saturation of the linear gain by the nonlinear losses. This theory predicts the appearance of SPP modes with the stationary values of the amplitudes reached above the threshold and reveals the crucial role of nonlinearity in shaping of the SPP spectra. We start our analysis from the time-independent Maxwell equations for TM waves:

$$\partial_{zz}E'_x - \partial_{zx}E'_z = -D_x, \quad (1)$$

$$\partial_{zx}E'_x - \partial_{xx}E'_z = D_z. \quad (2)$$

Here z is the coordinate along the interface and the x direction is orthogonal to it, and both are measured in the units of $1/k=\lambda_{\rm vac}/(2\pi)$, where $\lambda_{\rm vac}$ is the vacuum wavelength. For the constitutive relation we assume $\vec{D}=(\varepsilon+\gamma|\vec{E}'|^2)\vec{E}'$, where $\varepsilon=\varepsilon'+i\varepsilon''$ is the linear permittivity and $\gamma=\gamma'+i\gamma''$ is the nonlinear susceptibility. Below we use ε and γ with subscripts d and m when referring to the dielectric $(x>0)$ and the metal $(x<0)$, respectively. The metal is assumed linear, $\gamma_m=0$. Amplification in the dielectric is described using the two-level model with relatively small SPP intensities. Susceptibility of the two-level atoms is $\chi(\delta)=\alpha(\delta-i)/(\Gamma^2+\delta^2)$ [9], where δ is the detuning of the SPP frequency from the atomic resonance frequency and $\Gamma=(1+|\vec{E}'|^2/|E_*|^2)^{1/2}$ is the dimensionless intensity dependent linewidth. δ and Γ are both normalized to the physical transition linewidth. α is the dimensionless gain per unit length. We assume below that the SPP intensity is less than the saturation intensity $|E_*|^2$; a particular value of the latter depends on a material choice and is not important here. The effect of the nonlinear saturation of the linear gain discussed below should not be confused with and attributed to $|E_*|^2$.

We seek solutions of Eqs. (1) and (2) in the form $\vec{E}'(x,z)=E_*\vec{E}(x)\exp(i\beta z)$, where β is the propagation constant. Expanding Γ and χ into the Taylor series in $|\vec{E}|^2$ we find $\varepsilon_d=\varepsilon_b+\alpha(\delta+i)/(1+\delta^2)$, $\gamma_d=\alpha(i-\delta)/(1+\delta^2)^2$, where ε_b is the dielectric constant of the background material hosting the two-level atoms. For $\alpha>0$ we have linear gain and nonlinear absorption, which both are maximal at the line center $\delta=0$. We are not taking into account the metal dispersion, assuming that it is negligible relative to the dispersion introduced by the strong two-level resonance. Linear limit of the theory developed below admits arbitrary complex β's. However nonlinear results require the stationarity of the SPP intensities with respect to the propagation coordinate z, which is achieved due to balance between all the loss and gain mechanisms. Formally the balance condition is expressed as $\mathrm{Im}\,\beta=0$.

The exponentially decaying for $x<0$ solutions can be readily found, since the problem is linear inside the metal: $E_x=Be^{q_m x}$, $E_z=(iq_m/\beta)Be^{q_m x}$, $\mathrm{Re}(q_m)>0$. Here $q_m^2=\beta^2-\varepsilon_m$ and $|B|^2$ is the SPP intensity on the metal side of the interface. The system of equations we need to solve for $x>0$ is

$$\beta^2 E_x + i\beta\partial_x E_z = [\varepsilon_d + \gamma_d(|E_x|^2+|E_z|^2)]E_x, \qquad (3)$$

$$i\beta\partial_x E_x - \partial_{xx}E_z = [\varepsilon_d + \gamma_d(|E_x|^2+|E_z|^2)]E_z. \qquad (4)$$

The boundary conditions require continuity of E_z and D_x at $x=0$. Assuming that E_{x0}, E_{z0} are the field components on the dielectric side of the interface and knowing solutions inside the metal we express the boundary conditions using only the fields in the dielectric:

$$\beta\varepsilon_m E_{z0} = iq_m[\varepsilon_d + \gamma_d(|E_{x0}|^2+|E_{z0}|^2)]E_{x0}, \qquad (5)$$

Solving Eqs. (3) and (4) perturbatively under the assumption that nonlinear terms $\gamma_d|\vec{E}|^2$ are small we found

$$E_x = Ae^{-q_d x}\{1 + w_x\gamma_d|A|^2 e^{-2x\mathrm{Re}q_d} + O(|\gamma_d|^2)\}, \qquad (6)$$

$$E_z = \frac{q_d}{i\beta}Ae^{-q_d x}\{1 + w_y\gamma_d|A|^2 e^{-2x\mathrm{Re}q_d} + O(|\gamma_d|^2)\}. \qquad (7)$$

Here $q_d^2=\beta^2-\varepsilon_d$, $\mathrm{Re}(q_d)>0$, and $w_{x,y}$ are some constants not shown here. $|A|^2$ characterizes the SPP intensity on the dielectric side of the interface. Substituting the above solutions into Eq. (5), we find the dispersion law for SPPs accounting for losses in metal nonlinearity and gain in the dielectric

$$\varepsilon_m q_d + \varepsilon_d q_m = \gamma_d|A|^2F + O(|\gamma_d|^2). \qquad (8)$$

The constant F is given by

$$F \equiv \left(\frac{|q_d|^2}{\beta^2}+1\right) \times \frac{q_d\varepsilon_m + q_m\varepsilon_d + 2\mathrm{Re}(q_d)(\beta^2\varepsilon_m/\varepsilon_d - q_m q_d)}{4\mathrm{Re}(q_d)(\mathrm{Re}(q_d)+q_d)}. \qquad (9)$$

Dispersion of nonlinear SPPs in the absence of gain and loss has been previously derived, e.g., in [10].

For $|A|=0$, Eq. (8) transforms into a well-known linear dispersion law for SPPs, which is readily resolved with respect to β: $\beta=\beta_l\equiv\sqrt{(\varepsilon_m\varepsilon_d)/(\varepsilon_d+\varepsilon_m)}$. Practically, SPPs can be excited either directly by the two-level emitters (dye molecules, quantum dots, etc.) or externally coupled into the system, e.g., through a prism or a grating. Depending on the spatial variations of the emitter density and on the excitation type (optical, electric, or chemical pumping), the population inversion and hence the gain coefficient α may vary with the distance from the interface, see, e.g., [4]. We will not consider these effects in order to focus on the role of nonlinearity and to derive a closed analytical expression for the nonlinear SPP dispersion. To obtain physical estimates for our dimensionless calculations, we used $\varepsilon_b=1.8$ (polymer) and $\varepsilon_m=-15+i0.4$ (silver at $\lambda_{\mathrm{vac}}=530$ nm). For these parameters, without the resonant atoms ($\alpha=0$) the characteristic SPP propagation distance is $(k\,\mathrm{Im}\,\beta_l)^{-1}\simeq 30\ \mu$m.

If we neglect the effect of nonlinearity, the condition $\mathrm{Im}\,\beta_l=0$ corresponds to the lossless SPP propagation, and it is achieved at the gain threshold $\alpha=\alpha_0$: $\alpha_0=1/2\varepsilon_m''[(|\varepsilon_m|^2-2\varepsilon_m''\varepsilon_b\delta)-\{(|\varepsilon_m|^2-2\varepsilon_m''\varepsilon_b\delta)^2-4(\varepsilon_m'')^2(\varepsilon_b)^2(1+\delta^2)\}^{1/2}]$. As expected, the lowest gain $\alpha_0=\alpha_{\min}$ required for the lossless propagation happens at the exact resonance ($\delta=0$): $\alpha_{\min}\simeq 0.00575$. For example for $\alpha=1.5\alpha_{\min}$ and $2\alpha_{\min}$ the characteristic SPP gain length $(k\,\mathrm{Im}\,\beta_l)^{-1}\simeq 65\ \mu$m and 30 μm, respectively. Intensity of the nonstationary ($\mathrm{Im}\,\beta\neq 0$) SPPs in the linear case can be easily calculated: $I_l\sim e^{-2z\,\mathrm{Im}\,\beta_l}$. The typical dependence of I_l on δ is shown in Fig. 1(a) (line 2). Lines 2 and 3 in Fig. 1(b) show how the FWHM of $I_l(\delta)$ varies with the gain parameter α for two different propagation distances. One can see that the spectrum quickly narrows as the gain is increased but kept below the threshold ($\alpha<\alpha_{\min}$). Close to and above the threshold the narrowing continues but at a much slower pace. With the increase of the propagation distance and for the fixed gain, the spectrum also narrows [cf. lines 2 and 3 in Fig. 1(b)], since the spectral components of SPP modes near the line center are stronger amplified and hence become dominant.

The influence of nonlinear effects on the SPP propagation constant can be derived by solving Eq. (9) with respect to β and demanding $\mathrm{Im}\,\beta=0$. We assume that the right-hand side of Eq. (9) can be treated as a perturbation and find the following:

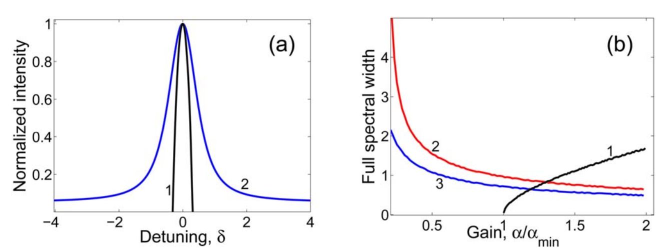

Fig. 1. (Color online) (a) Normalized SPP intensity as a function of detuning δ. Line 1 is the spectrum of stationary (saturated) SPP, $I_s(\delta)$: $\alpha=1.1\alpha_{\min}$. Line 2 is the spectrum of the amplified linear SPP, $I_l(\delta)$, below the threshold (propagation distance 60 μm, $\alpha=0.8\alpha_{\min}$). (b) FWHM of the SPP spectra versus gain. Line 1 is for the stationary saturated SPPs, I_s. Lines 2 (propagation distance 60 μm) and 3 (distance 100 μm) are for the nonstationary linear SPPs, I_l.

$$\beta = \beta_l + \beta_{nl}|A|^2, \quad \beta_{nl} \equiv \frac{\beta_l \gamma_d}{2\varepsilon_d^2} \frac{q_d(|q_d|^2 + \beta_l^2)}{\mathrm{Re}(q_d) + q_d}, \qquad (10)$$

where $q_{d,m}$ inside β_{nl} are calculated for $\beta=\beta_l$. Above the threshold ($\alpha > \alpha_{\min}$), the linear growth of the SPP intensity is saturated by the nonlinear absorption, and as a result the intensity quickly attains a stationary value. To calculate the spectral and other characteristics of the stationary (saturated) SPPs, we use Eq. (10) and impose the condition $\mathrm{Im}\,\beta=0$.

In the linear approximation, $\mathrm{Im}\,\beta_l=0$ implies $\alpha=\alpha_0$ (see above) and the corresponding real propagation constant is $\beta_l(\alpha_0)\equiv\beta_0$. Expanding β in Eq. (10) into the Taylor series in $(\alpha-\alpha_0)$, i.e., close to the threshold for a given δ, we find $\beta=\beta_0+(\alpha-\alpha_0)\partial_\alpha\beta_l+\beta_{nl}|A|^2$. Then $\mathrm{Im}\,\beta=0$ gives the intensity $I_s\equiv|A_s|^2$ of the stationary SPPs:

$$I_s = (\alpha - \alpha_0)\mathrm{Im}\,\partial_\alpha\beta_l/(-\mathrm{Im}\,\beta_{nl}), \qquad (11)$$

where $\partial_\alpha\beta_l$ and β_{nl} are calculated for $\alpha=\alpha_0$. Figure 2 shows dependencies of I_s on gain for several values of δ. Naturally, one can see that for higher gain, SPPs within the wider frequency interval around the resonance cross the threshold, leading to the gradual broadening of the spectra of the stationary SPPs [see line 1 in Fig. 1(b)].

Figure 1(a) compares the spectral intensity profiles of the linear SPPs below threshold and of the nonlinear saturated ones above the thresholds. Also, Fig. 1(b) compares the linewidth of these two SPP families. One can see that the spectra of stationary SPPs above the threshold are much narrower for small deviations from $\alpha_{\min}$ than spectra of the linear SPPs. For propagation distances of few gain lengths $(k\,\mathrm{Im}\,\beta_l)^{-1}$ (e.g., 100 μm) we shall expect that the SPPs should achieve their stationary saturated intensities. Therefore, the threshold crossover, if observed at sufficiently long distances, should be accompanied by a marked spectral narrowing, which has indeed been reported in the recent experiments [7]. Note the obvious qualitative agreement between our Figs. 1(a) and 2, and the experimental Figs. 2(a) and 2(b) in [7].

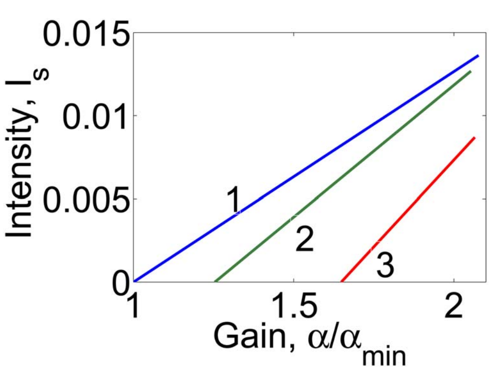

Fig. 2. (Color online) Dependence of the intensity of the stationary SPPs above the threshold on the gain parameter $\alpha/\alpha_{\min}$. Line 1 corresponds to $\delta=0$, line 2 corresponds to $\delta=0.4$, and line 3 corresponds to $\delta=0.8$.

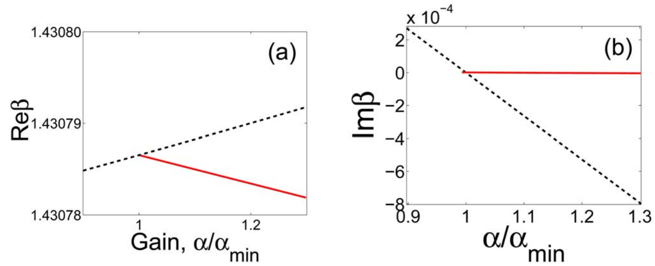

Fig. 3. (Color online) Dependence of the (a) real and (b) imaginary parts of the SPP propagation constants on the gain parameter for $\delta=0$. Solid lines correspond to the stationary nonlinear SPPs (β_s), and the dashed lines correspond to the linear SPPs (β_l).

For the propagation constant of the stationary saturated SPPs we thus have $\beta_s=\beta_0+(\alpha-\alpha_0)\mathrm{Re}\,\partial_\alpha\beta_l+I_s\,\mathrm{Re}\,\beta_{nl}$. Figure 3 compares real and imaginary parts of the above β_s with β_l. It shows that there exists a marked difference of the dependencies of the propagation constant from the gain parameter in the linear and nonlinear cases. We have independently cross-checked the SPP profiles and propagation constants using the numerical shooting method applied directly to the Maxwell equations. Good agreement between analytical and numerical results for $\alpha < 2\alpha_{\min}$ made it unnecessary to present the numerical results in the context of this Letter.

In summary, we have presented a theory of SPP amplification in the presence of nonlinear gain saturation as it happens in the real-world systems. We have calculated saturated intensities of SPPs above the amplification threshold and quantitatively described transition from the broad Lorentzian spectra of the nonstationary amplified SPPs to the narrow spectra of the stationary saturated SPPs above the threshold.

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References

1. D. J. Bergman and M. I. Stockman, Phys. Rev. Lett. **90**, 027402 (2003).
2. N. I. Zheludev, V. Prosvirnin, N. Papasimakis, and V. A. Fedotov, Nat. Photonics **2**, 351 (2008).
3. M. P. Nezhad, K. Tetz, and Y. Fainman, Opt. Express **12**, 4072 (2004).
4. I. De Lion and P. Berini, Phys. Rev. B **78**, 161401(R) (2008).
5. J. Seidel, S. Grafstrom, and L. Eng, Phys. Rev. Lett. **94**, 177401 (2005).
6. G. Winter, S. Wedge, and W. Barnes, New J. Phys. **8**, 125 (2006).
7. M. A. Noginov, G. Zhu, M. Mayy, B. A. Ritzo, N. Noginova, and V. A. Podolskiy, Phys. Rev. Lett. **101**, 226806 (2008).
8. M. Ambati, S. H. Nam, E. Ulin-Avila, D. A. Genov, G. Bartal, and X. Zhang, Nano Lett. **8**, 3998 (2008).
9. R. W. Boyd, *Nonlinear Optics* (Academic Press, 2003).
10. D. Mihalache, G. I. Stegeman, C. T. Seaton, E. M. Wright, R. Zanoni, A. D. Boardman, and T. Twardowski, Opt. Lett. **12**, 187 (1987).