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# Optical gain in surface plasmon nanocavity oscillators

**Chuan S. Liu and Vipin K. Tripathi*†**
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**Abstract.** An analytical model of optical gain is developed for three types of surface plasmon nano-oscillators: (i) a metal film with a gain medium nanostrip, (ii) a metal film, with nanohole, deposited on a layer of gain medium, and (iii) a nanoparticle coated with a gain medium. The operating frequency of the plasmon laser is close to surface plasmon resonance, hence the cavity size is strongly reduced. The evanescent field of the oscillator stimulates the electron–hole recombination in the gain medium, amplifying the cavity field. With electron–hole occupation probabilities in the relevant energy states in the gain medium just exceeding 50% each, the growth rate exceeds $10^{13}\ \mathrm{s}^{-1}$. The excited cavity mode acts as an oscillatory dipole to emit optical radiation.  [DOI: 10.1117/1.JNP.10.016015]

**Keywords:** spaser; nanolaser; optical gain; surface plasmons.

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## 1 Introduction

The surface plasma wave (SPW), supported by a conductor–dielectric interface, has an important property: as the wave frequency approaches the surface plasmon resonance, the wavelength and transverse extent of the mode resonantly shrink, confining optical frequency waves to nanodimensions, far below the diffraction limit prescribed for body waves.[1,2] This has led to the development of nanoelectronics and nano-devices into a major field[1–5] of research. Of significant importance is the development of nanolaser or spaser.[6–10] Noginov et al.[11] have recently operated a 531 nm spaser-based nanolaser employing 44 nm diameter nanoparticles with gold core and dye-doped silica shell. Lu et al.[12,13] have developed a multilayer plasminic nanolaser using smooth silver film with $SiO_2$ or $Al_2O_3$ nanolayer and gallium nitride nanorod deposited on it. The nanorod comprises a layer of indium gallium nitride that acts as gain medium. The theoretical formalisms of spaser are usually based on quantum density matrix approach.[6] However, some employ classical model due to its transparent simplicity. Kumar et al.[14] developed a classical analytical formalism of two-layer surface plasmon laser amplifier pumped by a forward biased p–n junction.

In this paper, we present theoretical analysis of optical gain in three surface plasmonic nano-oscillators, (i) a metal film with a nanostrip of gain medium, (ii) a nanohole in a metal film deposited on a gain layer,[15] and (iii) a nanoparticle coated with gain medium. These are the configurations experimentally studied in recent years and have promise for building arrays of nanoradiators with desired directivity and power. Here we pursue analytical treatment to reveal physics with some clarity which at times gets masked in numerical simulations.

The oscillators operate near the surface plasmon resonance frequency and the quality factor of the cavity is determined by the free electron collisional damping. In the first oscillator, nanostrip provides for lateral localization of the surface plasmons[16] by reducing the phase velocity of the SPW (as compared to the metal–vacuum interface). In the second oscillator, mode is localized around the nanohole. In the third case, space charge oscillations of electron sphere with respect to ion sphere provide a natural oscillator. The field of the

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oscillator mode stimulates electron–hole recombination in the gain medium that in turn amplifies the mode.

In Sec. 2, we study the optical gain in a metal-nanostrip oscillator. In Secs. 3 and 4, we consider the nanohole and nanoparticle oscillators. In Sec. 5, we discuss the results.

## 2 Metal Film—Nanostrip Spaser

Consider a metal film of free electron density $n_0^0$, electron effective mass $m$, and lattice dielectric constant $\varepsilon_L$. On top of it ($x > 0$) lies a nanostrip with $0 < y < W$, $0 < z < L$ and dielectric constant $\varepsilon_d$ (cf. Fig. 1). The thickness of the metal film is larger than the skin depth of the SPW, hence it can be treated like a semi-infinite medium ($x < 0$). Grooves are created in the metal at $z = 0$, $L$ for longitudinal localization of plasmons.

Had it been an infinite metal–dielectric interface, it would support a SPW with electric field[17]

$$\vec{E} = \vec{F}_1(x)e^{-i(\omega t - \vec{k}_{||}.\vec{r}_{||})}, \tag{1}$$

$$\vec{F}_1 = A\left(\hat{k}_{||} - \frac{ik_{||}}{\alpha_{\mathrm{I}}}\hat{x}\right)e^{\alpha_{\mathrm{I}}x} \quad \text{for } x < 0$$

$$= A\left(\hat{k}_{||} + \frac{ik_{||}}{\alpha_{\mathrm{II}}}\hat{x}\right)e^{-\alpha_{\mathrm{II}}x} \quad \text{for } x > 0$$

$$k_{||} = \frac{\omega}{c}\left(\frac{\varepsilon_{\mathrm{m}}\varepsilon_{\mathrm{d}}}{\varepsilon_{\mathrm{m}} + \varepsilon_{\mathrm{d}}}\right)^{1/2},$$

$$\alpha_{\mathrm{I}} = \frac{\omega}{c}\left(\frac{-\varepsilon_{\mathrm{m}}^2}{\varepsilon_{\mathrm{m}} + \varepsilon_{\mathrm{d}}}\right)^{1/2}, \quad \alpha_{\mathrm{II}} = \frac{\omega}{c}\left(\frac{-\varepsilon_{\mathrm{d}}^2}{\varepsilon_{\mathrm{m}} + \varepsilon_{\mathrm{d}}}\right)^{1/2}, \tag{2}$$

where $\vec{k}_{||} = k_y\hat{y} + k_z\hat{z}$, $\vec{r}_{||} = y\hat{y} + z\hat{z}$, $\varepsilon_{\mathrm{m}} = \varepsilon_L - (\omega_{\mathrm{p}}^2/\omega^2)(1 - i\nu/\omega)$ is the effective relative permittivity of the metal, $\omega_{\mathrm{p}} = (n_0^0 e^2/m\varepsilon_0)^{1/2}$ is the plasma frequency, $-e$ is the electron charge, $\nu$ is the electron collision frequency, $\varepsilon_0$ is the free space permittivity. SPW exists for $\omega < \omega_{\mathrm{R}}$ where

$$\omega_{\mathrm{R}} = \omega_{\mathrm{P}}/(\varepsilon_{\mathrm{m}} + \varepsilon_{\mathrm{d}})^{1/2}. \tag{3}$$

For a given $\omega$, $k_{||}$ increases with increasing $\varepsilon_{\mathrm{d}}$.

When one limits the lateral size $W$ of the dielectric layer, the parallel wave number of the SPW on the metal–vacuum interface is

$$k_{0||} = \frac{\omega}{c}\left(\frac{\varepsilon_{\mathrm{m}}}{\varepsilon_{\mathrm{m}} + 1}\right)^{1/2}. \tag{4}$$

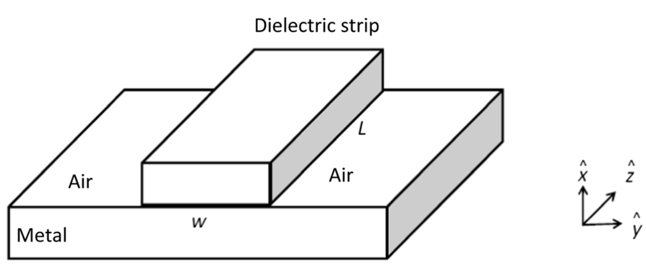


**Fig. 1** Schematic of nanostrip-loaded metal film. The nanostrip of length $L$, width $W$, and relative permittivity $\varepsilon_{\mathrm{d}}$ localizes the SPW. It contains a layer of gain medium that under optical pumping excites localized surface plasmons.

One may take the SPW to be propagating along $\hat{z}$ but with finite $k_y$. Outside the strip, i.e., on the metal–vacuum interface, the SPW would have the same $k_z$ as inside the strip. However, since $k_{0||} < k_{||}$, the SPW field outside the strip would be evanescent in $y$. One may take SPW to be localized in $y$ and $z$ with

$$k_y \approx \mathrm{n}_1\pi/W, \quad k_z \approx \mathrm{n}_2\pi/L, \tag{5}$$

where $n_1$ and $n_2$ are integers.

For the plasmon nano-oscillator, we may deduce the mode structure as follows. Take $H_x = 0$, $E_x \neq 0$. Assuming $x$, $t$ variations of fields as exp $(\alpha x - i\omega t)$ and using $\nabla \times \vec{E} = i\omega\mu_0\vec{H}$ and $\nabla \times \vec{H} = -\mathrm{i}\omega\varepsilon_0\varepsilon\vec{E}$ express $E_y$, $E_z$, $H_y$, $H_z$ in terms of $E_x$. In different regions one may take the $y$, $z$ variations of fields to be the same [with $k_y$, $k_z$ given by Eq. (5)], in compliance with the boundary conditions at $x = 0$ and allow $\alpha$ to have different values in different media. Thus we write in the region $0 < y < W$, $0 < z < L$

$$\begin{aligned}
&x < 0\\
&\quad E_x = A_1 F_x e^{\alpha_I x} e^{-i\omega t},\\
&\quad E_y, H_z = (\alpha_I, i\omega\varepsilon_0\varepsilon_\mathrm{m})\frac{k_y A_1}{k_{||}^2} F_y e^{\alpha_I x} e^{-i\omega t},\\
&\quad E_z, H_y = (\alpha_\mathrm{I}, \mathrm{i}\omega\varepsilon_0\varepsilon_\mathrm{m})\frac{k_z A_1}{k_{||}^2} F_z e^{\alpha_\mathrm{I} x} e^{-i\omega t},
\end{aligned} \tag{6}$$

$$\begin{aligned}
&x > 0\\
&\quad E_x = A_2 F_x e^{-\alpha_{II} x} e^{-i\omega t},\\
&\quad E_y, H_z = (-\alpha_\mathrm{II}, i\omega\varepsilon_0\varepsilon_\mathrm{d})\frac{k_y A_2}{k_{||}^2} F_y e^{-\alpha_\mathrm{II} x} e^{-i\omega t},\\
&\quad E_z, H_z = (-\alpha_{II}, -i\omega\varepsilon_0\varepsilon_\mathrm{d})\frac{k_z A_2}{k_{||}^2} F_z e^{-\alpha_\mathrm{II} x} e^{-i\omega t},
\end{aligned} \tag{7}$$

$$F_x = \sin(k_y y)\sin(k_z z), \quad F_y = \cos(k_y y)\sin(k_z z), \quad F_z = \sin(k_y y)\cos(k_z z),$$

$$\alpha_\mathrm{I}^2 = k_{||}^2 - \frac{\omega^2}{c^2}\varepsilon_\mathrm{m}, \quad \alpha_\mathrm{II}^2 = k_{||}^2 - \frac{\omega^2}{c^2}\varepsilon_\mathrm{d}.$$

Applying the continuity of $\varepsilon E_x$, $E_z$ at $x = 0$, we obtain $A_2 = A_1\varepsilon_\mathrm{d}/\ \varepsilon_\mathrm{m}$ and the dispersion relation

$$F_2 \equiv 1 + \alpha_\mathrm{I}\varepsilon_\mathrm{d}/\alpha_\mathrm{II}\varepsilon_\mathrm{m} = 0, \tag{8}$$

giving $k_{||}$ as in Eq. (2). Since $k_{||}$ is quantized [cf. Eq. (5)], we get discrete eigenfrequencies $\omega$. In the absence of collisions ($\nu = 0$), $F_2$ is real and so are the frequencies. For $\nu \neq 0$, $F_2$ has a finite imaginary part, $F_2 = F_{2\mathrm{r}}(\omega) + iF_{2i}$. Writing $\omega = \omega_r - i\Gamma$, we obtain

$$\omega_\mathrm{r}^2 = \frac{\omega_\mathrm{p}^2}{2\varepsilon_L}\left[1 + \left(1 + \frac{\varepsilon_L}{\varepsilon_\mathrm{d}}\right)\beta^2 - \left\{\left[1 + \left(1 + \frac{\varepsilon_L}{\varepsilon_\mathrm{d}}\right)\beta^2\right]^2 - 4\frac{\varepsilon_L}{\varepsilon_\mathrm{d}}\beta^2\right\}^{1/2}\right], \tag{9}$$

$$\Gamma = -\frac{F_{2i}}{(\partial F_{2\mathrm{r}}/\partial\omega)} = \frac{\nu}{2}\frac{\omega_\mathrm{p}^2}{\omega_\mathrm{r}^2}\frac{\varepsilon_\mathrm{d}}{\varepsilon_\mathrm{m}^2 + \varepsilon_\mathrm{L}\varepsilon_\mathrm{d}}, \tag{10}$$

where $\beta^2 = (\pi^2 c^2/\omega_\mathrm{p}^2 W^2)(n_1^2 + n_2^2 W^2/L^2)$. We have plotted in Fig. 2(a) the real frequency of a surface plasmon eigenmode as a function of normalized width of the strip for typical parameters. The frequency steadily falls off with the width. Figure 2(b) displays the variation of normalized

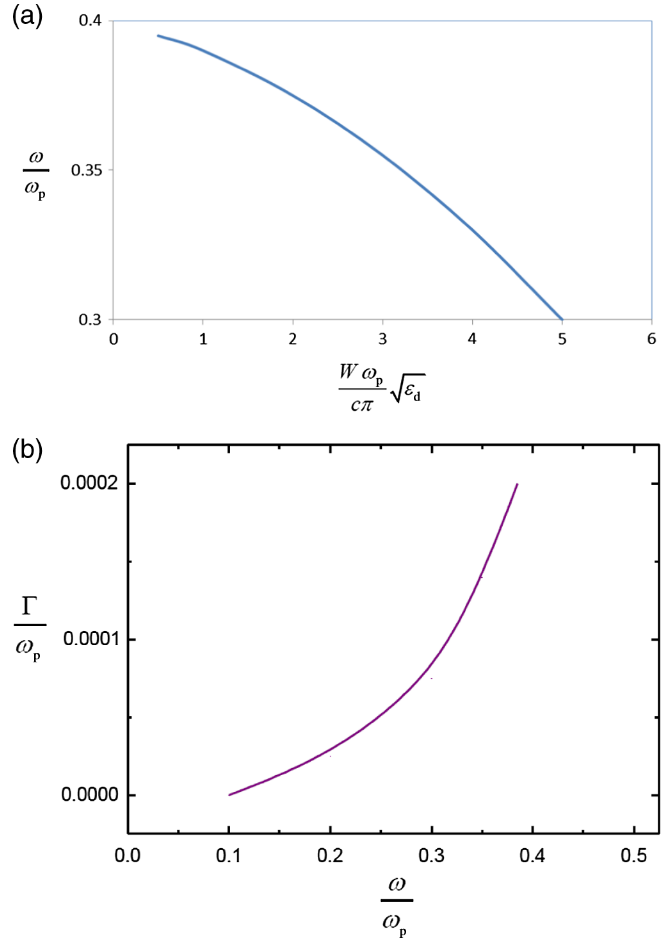


**Fig. 2** (a) Normalized eigenfrequency of the plasmon oscillator as a function of normalized width of the strip for $\varepsilon_L = 4$, $\varepsilon_d = 2.6$, $n_1 = 1$, $n_2 = 5$, $L/W = 3$. (b) Normalized damping rate as a function of normalized frequency for $\nu/\omega = 3 \times 10^{-4}$, $\varepsilon_L = 4$, $\varepsilon_d = 2.6$.

damping rate with normalized frequency. The damping increases rather rapidly as one approaches the surface plasmon resonance frequency.

The energy density of the electromagnetic fields of the plasmon resonator is

$$\begin{aligned} W_{\mathrm{EM}} &= \frac{\varepsilon_0}{4}\frac{\partial}{\partial\omega}(\omega\varepsilon)\vec{E}.\vec{E}^* + \frac{\mu_0}{4}\vec{H}.\vec{H}^* \\ &= \frac{\varepsilon_0}{4}\frac{A_1^2}{\alpha_\mathrm{I}^2}e^{2\alpha_\mathrm{I}x}\left[k_{||}^2\left(\in_\mathrm{L} + \frac{\omega_\mathrm{p}^2}{\omega^2}\right)\sin^2(k_y y)\sin^2(k_z z)\right. \\ &\quad + \left(\varepsilon_\mathrm{m} + \frac{2\omega_\mathrm{p}^2\alpha_\mathrm{I}^2}{k_{||}^2\omega^2}\right)\{k_z^2\sin^2(k_y y)\cos^2(k_z z) \\ &\quad \left. + k_y^2\cos^2(k_y y)\sin^2(k_z z)\}\right] \quad \text{for } x < 0, \\ &= \frac{\varepsilon_0\varepsilon_\mathrm{d}A_1^2}{4\alpha_\mathrm{II}^2}e^{-2\alpha_\mathrm{II}x}[k_{||}^2\sin^2(k_y y)\sin^2(k_z z) \\ &\quad + k_z^2\sin^2(k_y y)\cos^2(k_z z) + k_y^2\cos^2(k_y y)\sin^2(k_z z)] \quad \text{for } x > 0, \end{aligned} \tag{11}$$

where $\varepsilon = \varepsilon_\mathrm{m}$ for $x < 0$, $\varepsilon = \varepsilon_\mathrm{d}$ for $x > 0$. The total energy stored in the SPW is

$$E_{\mathrm{EM}} = \int_0^L \int_0^W \int_{-\infty}^{\infty} W_{\mathrm{EM}}\,\mathrm{d}x\,\mathrm{d}y\,\mathrm{d}z = \delta_1 A_1^2,$$

$$\delta_1 = \frac{\varepsilon_0 W L k_\parallel^2}{16}\left[\frac{\varepsilon_\mathrm{d}}{\alpha_\mathrm{II}^3} + \frac{1}{\alpha_\mathrm{I}^3}\left(\varepsilon_\mathrm{L} - \frac{\omega_\mathrm{p}^2}{\omega^2}\frac{\varepsilon_\mathrm{m}}{\varepsilon_\mathrm{d}}\right)\right]. \tag{12}$$

## 2.1 *Optical Gain*

Now we allow the dielectric strip to comprise a gain medium (e.g., an optically pumped semiconductor layer) of $x$-width $\Delta$, $y$-width $W$, and $z$-extent $L$, located at $x = \mathrm{d}$. In the conduction and valence bands, the density of states and occupation probability for electrons and holes (of effective masses $m_\mathrm{e}$, $m_\mathrm{h}$), respectively, are

$$\rho_\mathrm{e}(E_\mathrm{e}) = \frac{1}{4\pi^2}\left(\frac{2\ \mathrm{m_e}}{\hbar^2}\right)^{3/2} E_\mathrm{e}^{1/2}, \quad \rho_\mathrm{h}(E_\mathrm{h}) = \frac{1}{4\pi^2}\left(\frac{2m_\mathrm{h}}{\hbar^2}\right)^{3/2} E_\mathrm{h}^{1/2}, \tag{13}$$

$$f_\mathrm{e}(E_\mathrm{e}) = [1 + \exp((E_\mathrm{e} - E_\mathrm{Fe})/T)]^{-1}, \quad f_\mathrm{h}(E_\mathrm{h}) = [1 + \exp((E_\mathrm{h} - E_\mathrm{Fh})/\mathrm{T})]^{-1}, \tag{14}$$

where $E_\mathrm{e}$ and $E_\mathrm{h}$ are the electron and hole energies measured from the bottom of the conduction band upward and top of the value band downward, respectively, $E_\mathrm{Fe}$ and $E_\mathrm{Fh}$ are the Fermi energies related to electron and hole densities $n_\mathrm{e}$ and $n_\mathrm{h}$ and temperature $T$ (in energy units).

In a surface plasmon-induced emission process, an electron in energy state $E_\mathrm{e}$ in the conduction band goes to energy state $E_\mathrm{h}$ in the valence band, recombining with a hole and producing a plasmon of frequency

$$\omega = (E_\mathrm{e} + \mathrm{E_h} + \mathrm{E_g})/\hbar, \tag{15}$$

where $E_\mathrm{g}$ is the band gap. The $k$ vector of the plasmon is much smaller than that of the electron before or after the transition, hence in a direct band gap parabolic band semiconductor we may take

$$E_\mathrm{e} = \hbar^2\mathrm{k}^2/2m_\mathrm{e}, \quad E_\mathrm{h} = \hbar^2\mathrm{k}^2/2\ \mathrm{m_h}. \tag{16}$$

Equations (15) and (16) give the energy states that participate in the stimulated emission process

$$E_\mathrm{e} = (\hbar\omega - \mathrm{E_g})/(1 + m_\mathrm{e}/m_\mathrm{h}),$$

$$E_\mathrm{h} = (\hbar\omega - \mathrm{E_g})/(1 + m_\mathrm{h}/m_\mathrm{e}). \tag{17}$$

The rate of e–h recombination per unit volume per second is proportional to spectral energy density $u_\omega \mathrm{d}\omega = W_\mathrm{EM}\delta(\omega - \omega_\mathrm{m})\mathrm{d}\omega$, density of states $\rho_\mathrm{e}(E_\mathrm{e})$, the occupation probability $f_\mathrm{e}(E_\mathrm{e})$ of state $E_\mathrm{e}$, the probability of state $E_\mathrm{h}$ being vacant, $f_\mathrm{h}(E_\mathrm{h})$

$$R_\mathrm{E} = B\rho_\mathrm{e}(E_\mathrm{e})f_\mathrm{e}(E_\mathrm{e})f_\mathrm{h}(E_\mathrm{h})W_\mathrm{EM}\delta(\omega - \omega_\mathrm{m})\mathrm{d}\omega, \tag{18}$$

where $B$ is the Einstein's coefficient of stimulated emission and $\omega_\mathrm{m}$ is the eigenfrequency of the SPW mode. Similarly, the rate of transition of electrons from the valence band to conduction band per unit volume per second is

$$R_\mathrm{abs} = B\rho_\mathrm{e}(E_\mathrm{e})(1 - f_\mathrm{e}(E_\mathrm{e}))(1 - f_\mathrm{h}(E_\mathrm{h}))W_\mathrm{EM}\delta(\omega - \omega_\mathrm{m})\mathrm{d}\omega. \tag{19}$$

One may remember that $R_\mathrm{abs}$ is proportional to the density of states in the valence band $\rho_\mathrm{h}(E_\mathrm{h}) = (m_\mathrm{h}/m_\mathrm{e})\rho_\mathrm{e}(E_\mathrm{e})$. The net SPW energy produced per unit volume per second on integrating over $\omega$ is

$$P = \mathrm{B}\rho_\mathrm{e}(E_\mathrm{e})(f_\mathrm{e}(E_\mathrm{e}) + f_h(E_\mathrm{h}) - 1)\hbar\omega W_\mathrm{EM}, \tag{20}$$

where we have dropped the subscript m over $\omega$. Integrating it over the volume of the gain medium, we obtain the rate of energy gain by the SPW oscillator

$$\frac{d}{dt}E_{\mathrm{EM}} + 2\Gamma E_{\mathrm{EM}} = \delta_2 A_1^2, \tag{21}$$

$$\delta_2 = \frac{\varepsilon_0\varepsilon_{\mathrm{d}} k_{||}^2}{8\alpha_{\mathrm{I}}^2}\Delta W L\hbar\omega B e^{-2\alpha_{\mathrm{I}} d}.\rho_{\mathrm{e}}(E_{\mathrm{e}})[f_{\mathrm{e}}(E_{\mathrm{e}}) + f_{\mathrm{h}}(E_{\mathrm{h}}) - 1]. \tag{22}$$

Using Eqs. (12) and (21) we obtain

$$A_1^2 = A_{00}^2 e^{\gamma t},$$

$$\gamma = \delta_2/\delta_1 - 2\Gamma = \frac{2\alpha_1\Delta e^{-2\alpha_1 \mathrm{d}} B\rho_{\mathrm{e}}(E_{\mathrm{e}})\hbar\omega}{[\alpha_{\mathrm{I}}^3/\alpha_{\mathrm{II}}^3 + (\varepsilon_{\mathrm{L}} - \varepsilon_{\mathrm{m}}\omega_p^2/\omega^2\varepsilon_{\mathrm{d}})/\varepsilon_{\mathrm{d}}]}.[f_{\mathrm{e}}(E_{\mathrm{e}}) + f_{\mathrm{h}}(E_{\mathrm{h}}) - 1] - 2\Gamma. \tag{23}$$

$\gamma$ is the growth rate of SPW field energy. The threshold for the SPW growth is given as

$$f_{\mathrm{e}} + f_{\mathrm{h}} \geq 1 + \frac{[\alpha_{\mathrm{I}}^3/\alpha_{\mathrm{II}}^3 + (\varepsilon_{\mathrm{L}} - \varepsilon_{\mathrm{m}}\omega_p^2/\omega^2\varepsilon_{\mathrm{d}})/\varepsilon_{\mathrm{d}}]\nu\omega_{\mathrm{p}}^2\varepsilon_{\mathrm{m}}\varepsilon_{\mathrm{d}}}{2(\varepsilon_L\varepsilon_{\mathrm{d}} + \varepsilon_{\mathrm{m}}^2)\alpha_1\Delta e^{-2\alpha_1 d}\mathrm{B}\rho_{\mathrm{e}}(\varepsilon_{\mathrm{e}})\hbar\omega}. \tag{24}$$

This determines the electron and hole density threshold for the growth of surface plasmons.

The Einstein's coefficient $B$ is related to the Einstein's coefficient for spontaneous emission $A$ as $B = \pi A c^3/\omega^3$, hence

$$B\rho_{\mathrm{e}}(E_{\mathrm{e}})\hbar\omega = \frac{1}{4\pi}A\left(\frac{2m_{\mathrm{e}}c^2}{\hbar\omega}\right)^{3/2}\left(\frac{E_{\mathrm{e}}}{\hbar\omega}\right)^{1/2},$$

which is of the order of $10^8 A$. Typical value of $A$, the inverse e–h recombination time, is $\sim 10^7\ \mathrm{s}^{-1}$. Further, for spaser operating around $\omega \sim \omega_R$ , $\varepsilon_{\mathrm{m}} \sim -\varepsilon_{\mathrm{d}}$, $\alpha_{\mathrm{I}}/\alpha_{\mathrm{II}} = 1$. For silver, gallium nitride case $\varepsilon_{\mathrm{m}} \sim 4$, $\varepsilon_{\mathrm{d}} \sim 2.5$. Thus, when the occupation probabilities $f_{\mathrm{e}}(E_{\mathrm{e}})$ and $f_{\mathrm{h}}(E_{\mathrm{h}})$ are just greater than 0.5 each the threshold for the spaser is exceeded and the growth rate of the order of $10^{13}\ \mathrm{s}^{-1}$ is achieved.

## 3 Nanohole-Embedded Metal Film Spaser

Consider a metal film of thickness $d$ $(0 < z < d)$ and effective relative permittivity $\epsilon_{\mathrm{m}}$ given above. It comprises a transverse hole of radius $a$ and length $d$ with vacuum inside (cf. Fig. 3).

The cylindrical vacuum–metal interface supports an SPW with evanescent fields in $r$ and forward backward propagating solutions in $z$. A forward wave SPW field is

$$\vec{E} = \vec{F}(r)e^{-i(\omega t - k_z z)}, \tag{25}$$

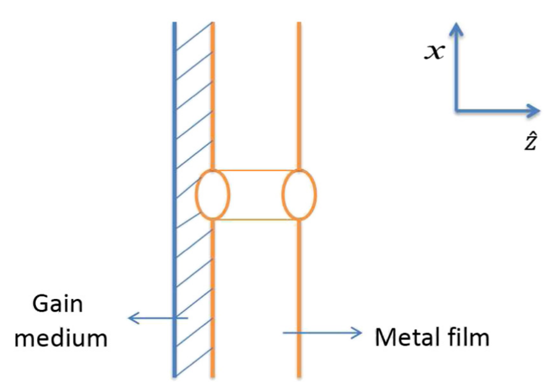


**Fig. 3** Schematic of a cylindrical hole drilled in a metal film deposited on a gain medium.

$$\vec{F} = A_1\left[\hat{z}I_0(\alpha_I r) - \hat{r}\frac{ik_z}{\alpha_I}I_1(\alpha_I r)\right] \quad \text{for } r < a$$

$$= A_2\left[\hat{z}K_0(\alpha_{II} r) + \hat{r}\frac{ik_z}{\alpha_{II}}K_1(\alpha_{II} r)\right] \quad \text{for } r > a,$$

where $\alpha_{\mathrm{I}} = (k_z^2 - \omega^2/c^2)^{1/2}$, $\alpha_{\mathrm{II}} = (k_z^2 - \omega^2\varepsilon_{\mathrm{m}}/c^2)^{1/2}$ and $I_0, K_0, I_1, K_1$ are the modified Bessel functions. The continuity of $E_z$, $\varepsilon E_r$ at $r = a$ gives the dispersion relation

$$\frac{I_1(\alpha_{\mathrm{I}}a)K_0(\alpha_{\mathrm{II}}a)}{I_0(\alpha_{\mathrm{I}}a)K_1(\alpha_{\mathrm{II}}a)} = -\frac{\alpha_{\mathrm{I}}}{\alpha_{\mathrm{II}}}. \tag{26}$$

The termination of the cylinder at $z = 0$, $d$ quantizes $k_z$ and gives standing wave solution to SPW. One may write

$r < a$

$$\begin{aligned}\vec{E} &= A_1\left[\hat{z}I_0(\alpha_{\mathrm{I}}r)\cos(k_z z) + \hat{r}\frac{k_z}{\alpha_{\mathrm{I}}}I_1(\alpha_{\mathrm{I}}r)\sin(k_z z)\right]e^{-i\omega t},\\ \vec{H} &= \hat{\varphi}\frac{\omega\varepsilon_0}{i\alpha_{\mathrm{I}}}A_1 I_1(\alpha_I r)\cos(k_z z)e^{-i\omega t},\end{aligned} \tag{27}$$

$r > a$

$$\begin{aligned}\vec{E} &= A_2\left[\hat{z}K_0(\alpha_{\mathrm{II}}r)\cos(k_z z) - \hat{r}\frac{k_z}{\alpha_{\mathrm{II}}}K_1(\alpha_{\mathrm{II}}r)\sin(k_z z)\right]e^{-i\omega t},\\ \vec{H} &= -\hat{\phi}\frac{\omega\varepsilon_0\varepsilon_{\mathrm{m}}}{i\alpha_{\mathrm{II}}}A_2 K_1(\alpha_{\mathrm{II}}r)\cos(k_z z)e^{-i\omega t},\end{aligned} \tag{28}$$

with $A_2 = A_1 I_0(\alpha_{\mathrm{I}}a)/K_0(\alpha_{\mathrm{II}}a)$ and $k_z = \pi/d$ for the fundamental mode. The dispersion relation, Eq. (26), gives the eigenfrequency and damping rate of the SPW mode.

The energy density of fields of the plasmon nano-oscillator is

$$\begin{aligned}W_{\mathrm{EM}} &= \frac{\varepsilon_0 A_1^2}{4\alpha_{\mathrm{I}}^2}\left[I_1^2(\alpha_{\mathrm{I}}r)\left(\frac{\omega^2}{c^2}\cos^2(k_z z) + k_z^2\sin^2(k_z z)\right) + \alpha_{\mathrm{I}}^2 I_0^2(\alpha_{\mathrm{I}}r)\cos^2(k_z z))\right] \quad \text{for } r < a\\ &= \frac{\varepsilon_0 A_2'^2}{4\alpha_{\mathrm{II}}^2}\left[K_0^2(\alpha_{\mathrm{II}}r)\alpha_{\mathrm{II}}^2\left(\varepsilon_L + \frac{\omega_{\mathrm{p}}^2}{\omega^2}\right)\cos^2(k_z z)\right.\\ &\quad \left. + K_1^2(\alpha_{\mathrm{II}}r)\left\{k_z^2\left(\varepsilon_L + \frac{\omega_{\mathrm{p}}^2}{\omega^2}\right)\sin^2(k_z z) + \frac{\omega^2}{c^2}\varepsilon_{\mathrm{m}}\cos^2(k_z z)\right\}\right] \quad \text{for } r > a.\end{aligned} \tag{29}$$

The total energy stored in the SPW is

$$E_{\mathrm{EM}} = \delta_1 A_1^2, \quad \delta_1 = \frac{\varepsilon_0 d}{8\alpha_{\mathrm{I}}^2}\left[G_1 + G_2\frac{\alpha_{\mathrm{I}}^2 I_0^2(\alpha_{\mathrm{I}}a)}{\alpha_{\mathrm{II}}^2 K_0^2(\alpha_{\mathrm{II}}a)}\right]$$

$$G_1 = \int_0^{\alpha_I a}\left[I_0^2(\xi) + \frac{1}{\alpha_{\mathrm{I}}^2}\left(\frac{\omega^2}{c^2} + \frac{\pi^2}{d^2}\right)I_1^2(\xi)\right]\xi \mathrm{d}\xi$$

$$\mathrm{G}_2 = \int_{\alpha_{\mathrm{II}}\mathrm{a}}^{\infty}\left[\left(\varepsilon_L + \frac{\omega_{\mathrm{p}}^2}{\omega^2}\right)K_0^2(\xi) + K_1^2(\xi)\left(k_{\mathrm{z}}^2\left(\varepsilon_L + \frac{\omega_{\mathrm{p}}^2}{\omega^2}\right) + \frac{\omega^2}{c^2}\varepsilon_{\mathrm{m}}\right)\right]\xi \mathrm{d}\xi. \tag{30}$$

## 3.1 *Optical Gain*

We allow a thin layer of gain medium of thickness $\Delta$ placed at $z = 0$. The net SPW energy produced per unit volume per second, $P$, is given by Eq. (20). The rate of energy gain of the SPW oscillator is given by Eq. (21) with

$$\delta_2 = B\rho_e(E_e)[f_e(E_e) + f_h(E_h) - 1]\hbar\omega \frac{\varepsilon_0 \Delta}{4\alpha_I^2} \cdot \left[G_3 + G_4 \frac{\alpha_I^2 I_0^2(\alpha_I a)}{\alpha_{II}^2 K_0^2(\alpha_{II} a)}\right], \tag{31}$$

$$\mathrm{G}_3 = \int_0^{\alpha_I a} \left[\mathrm{I}_0^2(\xi) + \left(\frac{\omega^2}{c^2 \alpha_I^2}\right) \mathrm{I}_1^2(\xi)\right] \xi d\xi$$

$$\mathrm{G}_4 = \int_{\alpha_{II} a}^{\infty} \left[\left(\varepsilon_L + \frac{\omega_p^2}{\omega^2}\right) K_0^2(\xi) + \left(\frac{\omega^2 \varepsilon_m}{c^2}\right) K_1^2(\xi)\right] \xi d\xi,$$

The linear damping rate $\Gamma$ is of the order of the one given by Eq. (10). Equation (21) gives $\mathrm{A}_I^2 = \mathrm{A}_{00}^2 \exp(\gamma \mathrm{t})$ with growth rate

$$\gamma = \delta_2/\delta_1 - 2\Gamma = \frac{2\Delta\hbar\omega}{d} B\rho_e(E_e)[f_e(E_e) + f_h(E_h) - 1] \cdot \frac{G_3 + G_4 \dfrac{\alpha_I^2 I_0^2(\alpha_I a)}{\alpha_{II}^2 K_0^2(\alpha_{II} a)}}{G_1 + G_2 \dfrac{\alpha_I^2 I_0^2(\alpha_I a)}{\alpha_{II}^2 K_0^2(\alpha_{II} a)}} - 2\Gamma. \tag{32}$$

The threshold value of $f_e(E_e) + f_h(E_h)$ above which the SPW grows is obtained by putting $\gamma = 0$ in Eq. (32). Above the threshold the growth rate is comparable to the earlier case.

## 4 Nanoparticle Spaser

Consider a metallic nanoparticle of radius $r_c$, free electron density $n_0^0$, electron effective mass $m$, and lattice permittivity $\varepsilon_L$. It is surrounded by a layer of gain medium of thickness $\Delta$ and dielectric constant $\varepsilon_d$ (cf. Fig. 4).

For a moment, let us take $\varepsilon_d = 1$ and give a displacement $\vec{\delta}$ to free electrons of the nanoparticles. This creates a space charge field $\vec{E}_s = (n_0^0 e/(\varepsilon_0(\varepsilon_L + 2)))\vec{\delta}$ in the overlap region of the free electron sphere and ion sphere. Each electron thus experiences a restoration force $-(m\omega_p^2/(\varepsilon_L + 2))\vec{\delta}$, where $\omega_p = (n_0^0 e^2/\varepsilon_0 m)^{1/2}$. The momentum loss per electron per second via collisions is $m\nu\vec{\nu}$ where $\nu$ is the collision frequency and $\vec{\nu}$ is the drift velocity of electrons. Thus the equation of motion for an electron is[18]

$$\frac{d^2}{dt^2}\vec{\delta} + \nu \frac{d\vec{\delta}}{dt} + \frac{\omega_p^2}{(\varepsilon_L + 2)}\vec{\delta} = 0. \tag{33}$$

Presuming a solution $\vec{\delta} = \vec{\mathrm{A}}_1 \exp(-\mathrm{i}\omega \mathrm{t})$ we get

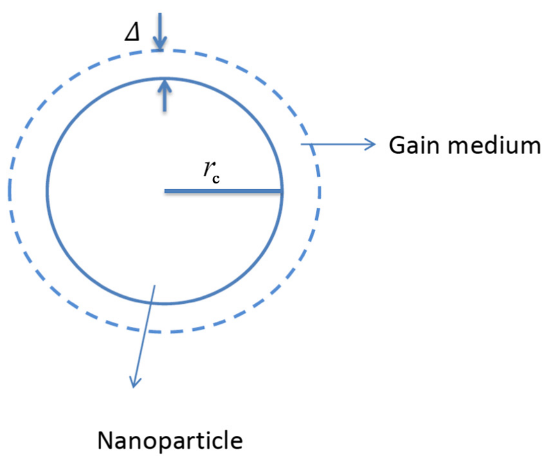


**Fig. 4** A nanoparticle of radius $r_c$ covered by a thin layer of gain medium of thickness $\Delta$.

$$\omega^2 + i\nu\omega - \frac{\omega_p^2}{(\varepsilon_L + 2)} = 0, \tag{34}$$

giving $\omega = \omega_r - i\Gamma$,

$$\omega_r = \omega_p / \sqrt{(\varepsilon_L + 2)},$$

$$\Gamma \cong \nu/2, \tag{35}$$

where we have assumed $\nu^2 \ll \omega^2$. Thus the nanoparticle is a natural oscillator of frequency $\omega_p/\sqrt{(\varepsilon_L + 2)}$ and quality factor $Q = \omega_p/(\nu\sqrt{(\varepsilon_L + 2)})$.

When we allow $\varepsilon_d$ to have arbitrary value, the Poisson's equation, governing the space charge field $\nabla.\varepsilon\nabla\phi = 0$, on writing $\vec{E} = -\nabla\phi$, gives the solution (for $\vec{E}||\hat{z}$)

$$\phi = C_1 r \cos\theta \quad \text{for } r < r_c, \quad \phi = C_2 \cos\theta/r^2 \quad \text{for } r > r_c.$$

Demanding the continuity of $\phi$, $\varepsilon\partial\phi/\partial r$ at $r = r_c$ we obtain

$$\varepsilon_m + 2\varepsilon_d = 0.$$

Using the value of $\varepsilon_m$ given below Eq. (2), we obtain the frequency given by Eq. (35) with $\varepsilon_L + 2$ replaced by $\varepsilon_L + 2\varepsilon_d$. This is the same result obtained by Bergman and Stockman[19] and Noginov et al.[20] for the fundamental mode.

The field of the nanoparticle [of dipole moment $-e\vec{\delta}(4\pi n_0^0 r_c^3/3)$] as seen by the gain layer is

$$\vec{E} = \frac{n_0^0 e r_c^3}{3\varepsilon_0\varepsilon_d r^3}\left(\vec{\delta} - 3\frac{\vec{\delta}.\vec{r}}{r^2}\vec{r}\right). \tag{36}$$

Outside the gain layer ($r > r_0 + \Delta$) the field is given by the above expression with $\varepsilon_d$ replaced by 1.

The energy stored in the oscillator is

$$E_{EM} = \frac{\varepsilon_0}{4}\int \frac{\partial}{\partial\omega}(\varepsilon\omega)|E|^2 dV = \delta_1 A_1^2, \tag{37}$$

$$\delta_1 = \frac{4\pi}{3} n_0^0 r_c^3 \frac{m\omega_p^2}{18}(1 + 2/\varepsilon_L),$$

where the volume integration has been carried out from inside the particle to the entire outside.

The oscillator field induces e–h recombination producing electromagnetic energy per second as given by Eq. (21) with $\Gamma$ given by Eq. (35) and

$$\delta_2 = \frac{2\pi}{9} n_0^0 m\omega_p^2 \Delta r_c^3 \hbar\omega B\rho_e(E_e)\cdot(f_e(E_e) + f_h(E_h) - 1). \tag{38}$$

The growth rate of the SPW turns out to be

$$\gamma = \delta_2/\delta_1 - 2\Gamma \quad = \frac{3\Delta\hbar\omega B}{r_c(1 + 2/\varepsilon_L)}\rho_e(E_e)[f_e(E_e) + f_h(E_h) - 1] - 2\Gamma. \tag{39}$$

For $B\rho_e(E_e)\hbar\omega \sim 10^{17}$ s$^{-1}$, $\Delta/r_c \sim 10^{-2}$, and $f_e(E_e)$, $f_h(E_h)$. One percent above the 50% occupation probability, the growth rate $\gamma \sim 10^{13}$ s$^{-1}$.

## 5 Discussion

The surface plasmon eigenmode in the vicinity of SPW resonance, $\omega \sim \omega_R$ is strongly localized near the metal–dielectric interface with $\alpha_I/\alpha_{II} \sim 1$ and has resonantly short wavelength. The damping rate of the mode, however, shows no resonant enhancement. A gain medium, within an SPW wavelength from the boundary sustaining SPW, excites the SPW eigenmode. Usually one employs optical pumping to achieve $f_e(E_e) + f_h(E_h) > 1$, the condition equivalent of population inversion. For electron and hole occupation probabilities of relevant energy states in the conduction and valence bands exceeding 0.5 each by 1% the growth rate is of the order of $10^{13}\ s^{-1}$.

In the case of metal film loaded with a gain medium nanostrip, frequency can be tuned by varying the permittivity and width of the strip. A drastic reduction in operating frequency can be achieved by shrinking the thickness of the metal film that lowers the SPW resonance frequency.[21] However, the present analysis is not valid in that case.

The nanohole oscillator has a gain comparable to the above oscillator at the same level of pumping power flux density. The SPW eigenmode acts as an oscillating dipole and emits far-field radiation. An array of such holes would give a well-collimated beam.

The oscillator comprising a nanoparticle coated with an optically pumped gain medium has operating frequency independent of the radius of the particle. It depends only on its free carrier density, lattice permittivity, and dielectric constant of the surrounding medium. Noginov et al.[20] have reported compensation of loss in metal nanoparticles oscillator by gain in interfacing rhodemine 6G dye. With emission cross section of R6G $q \approx 4 \times 10^{-16}\ cm^{-2}$ and density of active molecules $n_s = 2 \times 10^{18}\ cm^{-3}$ (corresponding to one molecule per nanoparticles of radius 5 nm) the local gain turns out to be $k_i \approx 10^3\ cm^{-1}$, corresponding to temporal growth $k_i c/\eta \approx 10^{13}\ s^{-1}$ which is comparable to our case where e–h recombination leads to growth of plasmons when $f_e \approx 0.5$, i.e., $n_e \approx 10^{18}\ cm^{-3}$. The quality factor $Q$ of the oscillators estimated here due to collisional losses appears to be higher than that reported experimentally. Photoabsorption via interband transitions appears to dominate collisional losses. These losses are equivalent to enhanced collision frequency of electrons.

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