Supporting Information:

Plasmonic Mirrorless Optical Parametric Oscillator

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Derivation of nonlinearly-coupled plasmon equations

The counter-propagating plasmonic waves can be described (under a quasi-continuous-wave approximation) by

$$\mathbf{E}_{\boldsymbol{s}} = A_s(x)\boldsymbol{\mathcal{E}}_{\boldsymbol{s}}(z)e^{i(k_s x - \omega_s t)} + C.C. \tag{S1}$$

and

$$\mathbf{E}_{\boldsymbol{i}} = A_i(x)\boldsymbol{\mathcal{E}}_{\boldsymbol{i}}(z)e^{i(-k_i x + \omega_i t)} + C.C. \tag{S2}$$

for the signal and idler waves, respectively. Here, $k_{s/i} = \frac{\omega_{s/i}}{c}\sqrt{\frac{\epsilon_m}{1+\epsilon_m}}$ are the in-plane wavenumbers of the plasmons, A_s and A_i represent their evolving amplitudes, $\boldsymbol{\mathcal{E}}_{\boldsymbol{s}}$ and $\boldsymbol{\mathcal{E}}_{\boldsymbol{i}}$ represent their envelopes, which are given by

$$\boldsymbol{\mathcal{E}}_{\boldsymbol{s/i}} = \begin{cases} \left[\pm\hat{\mathbf{x}} + i\sqrt{-\epsilon_m}\hat{\mathbf{z}}\right]e^{-\kappa_d^{s/i}z} & z > 0 \\ \left[\pm\hat{\mathbf{x}} - i\sqrt{-1/\epsilon_m}\,\hat{\mathbf{z}}\right]e^{+\kappa_m^{s/i}z} & z < 0 \end{cases} \tag{S3}$$

with $\kappa_d^{s/i} = \frac{\omega_{s/i}}{c}\sqrt{-\frac{1}{1+\epsilon_m}}$ and $\kappa_m^{s/i} = \frac{\omega_{s/i}}{c}\sqrt{-\frac{\epsilon_m^2}{1+\epsilon_m}}$ (such that $\nabla \cdot \mathbf{E}_{\boldsymbol{s/i}} = 0$ on each side of the metal-air interface), and $C.C.$ stands for complex conjugate. The incoming pump beam is taken as a free, p-polarized plane-wave, of the form

$$\mathbf{E}_{\boldsymbol{p}} = A_p\left[\cos\theta_p\hat{\boldsymbol{x}} + \sin\theta_p\hat{\boldsymbol{z}}\right]e^{i\left(\mathbf{k}_{\boldsymbol{p}}\cdot\boldsymbol{r}-\omega_p t\right)} + C.C. \qquad \text{(S4)}$$

with $\mathbf{k}_{\boldsymbol{p}} = k_p\left(\sin\theta_p\hat{\boldsymbol{x}} - \cos\theta_p\hat{\boldsymbol{z}}\right)$. Inside the metal, the waves interact through the third order nonlinearity of the metal. This interaction produces two polarization terms, at the signal and idler frequencies, which are given by

$$\mathbf{P}_{\boldsymbol{s}}^{\boldsymbol{NL}} = \epsilon_0\boldsymbol{\chi}^{(\mathbf{3})}\mathbf{E}_{\boldsymbol{p}}\mathbf{E}_{\boldsymbol{p}}\mathbf{E}_{\boldsymbol{i}}^* \qquad \text{(S5a)}$$

$$\mathbf{P}_{\boldsymbol{i}}^{\boldsymbol{NL}} = \epsilon_0\boldsymbol{\chi}^{(\mathbf{3})}\mathbf{E}_{\boldsymbol{p}}\mathbf{E}_{\boldsymbol{p}}\mathbf{E}_{\boldsymbol{s}}^* \qquad \text{(S5b)}$$

where $\boldsymbol{\chi}^{(\mathbf{3})}$ is the third-order susceptibility tensor of the metal film at the appropriate frequency. Below the metal-air interface ($z < 0$), the transmitted pump field can be written as $\mathbf{E}_{\boldsymbol{p}} = A_p\boldsymbol{\mathcal{E}}_{\boldsymbol{p}}e^{i\left(\mathbf{k}_{\boldsymbol{p}}\cdot\boldsymbol{r}-\omega_p t\right)} + C.C.$, with $\boldsymbol{\mathcal{E}}_{\boldsymbol{p}}$ given by

$$\boldsymbol{\mathcal{E}}_{\boldsymbol{p}} = \left[\left(1-r_p\right)cos\theta_p\hat{\boldsymbol{x}} + \frac{\left(1+r_p\right)}{\epsilon_m\left(\omega_p\right)}sin\theta_p\hat{\boldsymbol{z}}\right]e^{+\kappa_p z} \qquad \text{(S6)}$$

where r_p is the reflection coefficient of the p-polarized pump and $\kappa_p = \frac{\omega_p}{c}\sqrt{sin^2\theta_p - \epsilon_m}$. Assuming that the metal is polycrystalline and therefore exhibits isotropic nonlinearity, the nonlinear terms contributing to the FWM processes are $\chi_{ijij}^{(3)}$, $\chi_{ijji}^{(3)}$, $\chi_{iijj}^{(3)}$ and $\chi_{iiii}^{(3)} = \chi_{ijij}^{(3)} + \chi_{ijji}^{(3)} + \chi_{iijj}^{(3)}$. Since the two pump fields are identical we take $\chi_{ijij}^{(3)} = \chi_{iijj}^{(3)}$, and use the relation[1] $\chi_{ijji}^{(3)}/\chi_{ijij}^{(3)} = 4$. Using these terms, and taking into account that all of the waves are p-polarized, the nonlinear polarization is given by

$$\mathbf{P}_{\boldsymbol{s/i}}^{\boldsymbol{NL}} = \epsilon_0\chi^{(3)}A_p^2A_{s/i}^*\boldsymbol{\eta}\begin{pmatrix}\mathcal{E}_{i/s,x}^*\\0\\\mathcal{E}_{i/s,z}^*\end{pmatrix}e^{i\left[\left(2k_{\mathrm{p}}\sin\theta_p \pm k_{i/s}\right)x-\omega_{s/i}t\right]} \qquad \text{(S7)}$$

where the matrix $\boldsymbol{\eta}$ is given by

$$\boldsymbol{\eta} = \begin{pmatrix} 6\mathcal{E}_{p,x}^2 + 4\mathcal{E}_{p,z}^2 & 0 & 2\mathcal{E}_{p,x}\mathcal{E}_{p,z} \\ 0 & 0 & 0 \\ 2\mathcal{E}_{p,x}\mathcal{E}_{p,z} & 0 & 4\mathcal{E}_{p,x}^2 + 6\mathcal{E}_{p,z}^2 \end{pmatrix} \tag{S8}$$

Each one of the surface plasmon modes obeys an inhomogeneous wave equation, with the corresponding nonlinear polarization term acting as a source, such that

$$\left[\boldsymbol{\nabla}^2 - \frac{\epsilon_{d/m}}{c^2}\frac{\partial^2}{\partial t^2}\right]\mathbf{E}_{\boldsymbol{s/i}} = \mu_0 \frac{\partial^2}{\partial t^2}\mathbf{P}_{\boldsymbol{s/i}}^{\boldsymbol{NL}} \tag{S9}$$

Under phase-matching conditions (equations 1, 2), we may write the nonlinear polarization terms as

$$\mathbf{P}_{\boldsymbol{s/i}}^{\boldsymbol{NL}}(\boldsymbol{x},\boldsymbol{z}) = \epsilon_0 \chi^{(3)} A_p^2 A_{s/i}^* \boldsymbol{\mathcal{P}}_{\boldsymbol{s/i}}^{\boldsymbol{NL}} e^{i\left(\pm k_{s/i}x - \omega_s t\right)} \tag{S10}$$

with $\boldsymbol{\mathcal{P}}_{\boldsymbol{s/i}}^{\boldsymbol{NL}}(z) = \boldsymbol{\eta}\boldsymbol{\mathcal{E}}_{\boldsymbol{i/s}}^*$. In order to describe the evolution of the signal and idler amplitudes, we follow a procedure similar to Ref. 2. We assume that the amplitudes of the signal and idler are slowly-varying, such that $\left|\frac{\partial^2 A_{s/i}}{\partial x^2}\right| \ll \left|k_{s/i}\frac{\partial A_{s/i}}{\partial x}\right|$, and plug the plasmonic waves, given by equations S1-S2, into the wave equation S9, which yields

$$2ik_s\boldsymbol{\mathcal{E}}_{\boldsymbol{s}}\frac{\partial A_s}{\partial x} = -\frac{\omega_s^2}{c^2}\chi^{(3)}A_p^2 A_i^* \boldsymbol{\mathcal{P}}_{\boldsymbol{s}}^{\boldsymbol{NL}} \tag{S11a}$$

$$-2ik_i\boldsymbol{\mathcal{E}}_{\boldsymbol{i}}\frac{\partial A_i}{\partial x} = -\frac{\omega_i^2}{c^2}\chi^{(3)}A_p^2 A_s^* \boldsymbol{\mathcal{P}}_{\boldsymbol{i}}^{\boldsymbol{NL}} \tag{S11b}$$

Finally, we take the scalar product of equation S11a with $\boldsymbol{\mathcal{E}}_{\boldsymbol{s}}^*$ and equation S11b with $\boldsymbol{\mathcal{E}}_{\boldsymbol{i}}^*$, integrate over *z*, and replace A_p^2 with the pump intensity, to get a set of (linear) coupled equations for the amplitudes A_s and A_i:

$$\frac{\partial A_s}{\partial x} = ig_s A_i^* \tag{S12a}$$

$$\frac{\partial A_i}{\partial x} = -ig_i A_s^* \tag{S12b}$$

with the coupling constants being

$$g_{s/i} = \frac{\pi^2}{\lambda_{s/i}^2 k_{s/i}\epsilon_0 c}\frac{\beta_{s/i}}{\alpha_{s/i}}\chi^{(3)} I_p \tag{S13}$$

where $\lambda_{s/i} = 2\pi c/\omega_{s/i}$, $\alpha_{s/i} = \int_{-\infty}^{\infty}\left|\boldsymbol{\mathcal{E}}_{\boldsymbol{s/i}}\right|^2 dz$ and $\beta_{\boldsymbol{s/i}} = \int_{-\infty}^{\infty}\boldsymbol{\mathcal{P}}_{\boldsymbol{s/i}}^{\boldsymbol{NL}} \cdot \boldsymbol{\mathcal{E}}_{\boldsymbol{s/i}}^{*} dz$. Equation S12 may be further simplified by defining the normalized amplitudes $\mathrm{a_s} = \mathrm{A_s}/\sqrt{\mathrm{g_s}}$ and $\mathrm{a_i} = \mathrm{A_i}/\sqrt{\mathrm{g_i}}$, which yields

$$\frac{\partial a_s}{\partial x} = iGa_i^* \tag{S14a}$$

$$\frac{\partial a_i}{\partial x} = -iGa_s^* \tag{S14b}$$

with $G = \sqrt{g_s g_i} = \frac{\pi^2}{\lambda_s\lambda_i\epsilon_0 c}\sqrt{\frac{\beta_s\beta_i}{k_s k_i \mathrm{\alpha_s \alpha_i}}}\chi^{(3)} I_p$.

The boundary conditions of the counter propagating geometry are set by the input amplitudes on opposite sides for the signal and idler, such that $a_s(0) = a_{s0}$ and $a_i(L) = a_{i0}$, where L is the interaction length. This, in turn, is determined by the extent of the pump beam and the walk-off distance of the plasmons, which depends on the pulse duration time. Under these boundary conditions, the solution to equations S14 is given by

$$a_s(x) = \frac{\mathrm{a_{s0}}}{cos(GL)}cos[G(L-x)] + \frac{i\mathrm{a_{i0}^*}}{cos(GL)}sin(Gx) \tag{S15a}$$

$$a_i(x) = \frac{ia_{i0}}{cos(GL)}cos(Gx) + \frac{i\mathrm{a_{s0}^*}}{cos(GL)}sin[G(L-x)] \tag{S15b}$$

Similar to the traditional MOPO, these solutions diverge when GL aproaches $\pi/2$, signifying the onset of instability and the spontaneous oscillations of the signal and idler plasmons. This condition defines the critical pump intensity for the plasmonic parametric oscillations, which is

$$I_c = \frac{\lambda_s\lambda_i\epsilon_0 c}{2\pi\chi^{(3)}L}\sqrt{\frac{k_s k_i \mathrm{\alpha_s\alpha_i}}{\beta_s\beta_i}} \tag{S16}$$

Threshold intensity with plasmon absorption

The equations for the coupled plasmons (S14) can be modified to include the plasmon propagation losses, by adding a linear absorption term to propagation equations of the plasmons waves, such that

$$\frac{da_s}{dx} = iGa_i^* - k_s''a_s \tag{S17a}$$

$$\frac{da_i}{dx} = -iGa_s^* + k_i''a_i \tag{S17b}$$

with $k_{s/i}''$ being the imaginary parts of the plasmon wavenumbers. Similarly to equation S14, the solution of these coupled equations exhibits instability with diverging output amplitudes at a critical value of the nonlinear coupling, given by the condition[3]

$$cos\varphi + \frac{(k_s''+k_i'')L}{2\varphi} sin\varphi = 0 \tag{S18}$$

where $\varphi = \left[G^2 - \frac{(k_s''+k_i'')^2}{4}\right]^{1/2} L$. Notice that in the absence of losses, this condition reduces to $\varphi = GL = \pi/2$, as expected. With the losses included, equation S18 can be solved numerically for φ, from which the modified value of the threshold intensity can be calculated. It is easy to see that the inclusion of losses raises the critical value of φ above $\pi/2$. This, in effect, will increase the threshold intensity. We therefore define the threshold factor f as

$$f = \frac{GL}{\pi/2} = \frac{2}{\pi}\sqrt{\varphi^2 + \frac{(k_s''+k_i'')^2}{4} L^2} \tag{S19}$$

which, since G scales linearly with the pump intensity, represents the ratio between the threshold intensity with the losses included and the threshold intensity for the ideal, lossless case. As can be seen in Figure S1 (calculated with $L = 50\,\mu m$), in most of the existence range this factor is in the range of 1-2, indicating that the losses do not increase the threshold intensity dramatically. On the other hand, at wavelengths below 360 nm (corresponding to energies above ~ 3.5 eV), for which the plasmon propagation distance is significantly smaller than L, the threshold intensity rapidly increases due to the strong absorption of the interacting plasmons.

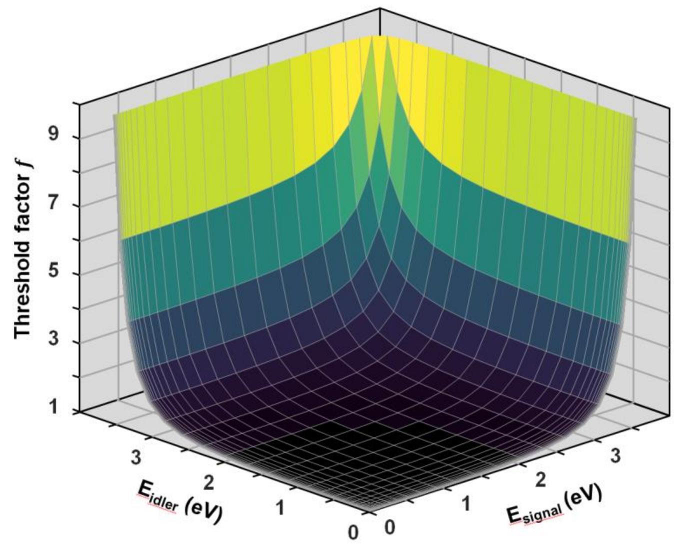

Figure S1. Threshold factor f as calculated from equations S18-S19 for a silver-air interface and with an interaction length of $L = 50\ \mu m$.

References

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