## Supplementary Information

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# Supplementary Information for “Emission dynamics and spectrum of a nanoshell-based plasmonic nanolaser spaser”

## 1 Extended Calculations

We here present all the extended analytical calculations that support the results presented in the main text.

### 1.1 Metal and Gain Medium Description

We start by the description of the materials composing the nanoshell.

Please note that all fields written without a tilde in this section correspond to real-valued quantities (measurable in the physical world), while fields with a tilde represent the corresponding complex amplitudes.

**1. Metal**

We begin by describing how the field $\mathbf{E}_\mathrm{m}(\mathbf{r},t)$, where $\mathbf{r}$ is the spatial coordinate with its origin at the particle center and $t$ is time, interacts with the electrons in the metallic nanoshell. This interaction is modeled using Drude's free-electron model:

$$\frac{d^2\mathbf{d}}{dt^2}+2\gamma\frac{d\mathbf{d}}{dt}=\frac{e}{m_\mathrm{e}}\mathbf{E}_\mathrm{m}, \tag{S.1}$$

where $\mathbf{d}$ represents the displacement of the electron cloud from its equilibrium position, $m_\mathrm{e}$ and $e$ are the electron mass and charge, respectively, and $\gamma$ is the ionic collision friction coefficient. We can then define the collective polarization produced by this displacement as:

$$\mathbf{\Pi}_\mathrm{m}=n_\mathrm{e}e\mathbf{d}, \tag{S.2}$$

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here $n_{\mathrm{e}}$ is the electron density in the metal. Substituting expression S.2 into S.1, and considering that the plasma frequency is given by

$$\omega_{\mathrm{p}}^2 = \frac{n_{\mathrm{e}} e^2}{\epsilon_0 m_{\mathrm{e}}}, \tag{S.3}$$

we can finally obtain the equation for the time evolution of $\mathbf{\Pi}_{\mathrm{m}}$:

$$\frac{d^2\mathbf{\Pi}_{\mathrm{m}}}{dt^2} + 2\gamma\frac{d\mathbf{\Pi}_{\mathrm{m}}}{dt} = \epsilon_0\omega_{\mathrm{p}}^2\mathbf{E}_{\mathrm{m}}. \tag{S.4}$$

$\mathbf{\Pi}_{\mathrm{m}}$ represents the dynamic component of the polarization in the metal. The total polarization experienced by the metal also includes the passive contribution from the ionic lattice:

$$\mathbf{P}_{\mathrm{m}} = \epsilon_0\chi_\infty\mathbf{E}_{\mathrm{m}} + \mathbf{\Pi}_{\mathrm{m}}.$$

Within the rotating wave approximation, the electric field and polarizations can be written in the following form:

$$\mathbf{E}_{\mathrm{m}}(t) = \frac{1}{2}\left[\tilde{\mathbf{E}}_{\mathrm{m}}(t)e^{-i\omega t} + \tilde{\mathbf{E}}^*_{\mathrm{m}}(t)e^{i\omega t}\right] \tag{S.5}$$

$$\mathbf{\Pi}_{\mathrm{m}}(t) = \frac{1}{2}\left[\tilde{\mathbf{\Pi}}_{\mathrm{m}}(t)e^{-i\omega t} + \tilde{\mathbf{\Pi}}^*_{\mathrm{m}}(t)e^{i\omega t}\right] \tag{S.6}$$

$$\mathbf{P}_{\mathrm{m}}(t) = \frac{1}{2}\left[\tilde{\mathbf{P}}_{\mathrm{m}}(t)e^{-i\omega t} + \tilde{\mathbf{P}}^*_{\mathrm{m}}(t)e^{i\omega t}\right], \tag{S.7}$$

where $\tilde{\mathbf{E}}_{\mathrm{m}}(t)$, $\tilde{\mathbf{\Pi}}_{\mathrm{m}}(t)$, and $\tilde{\mathbf{P}}_{\mathrm{m}}(t)$ represent a slow dependency on time (over times much slower than $1/\omega$).

If we now substitute expressions S.5-S.7 into equation S.4 and average over fast time variations (times of order $1/\omega$), we can finally obtain the time evolution equation for the dynamic part of the polarization in the metal region:

$$\frac{d\tilde{\mathbf{\Pi}}_{\mathrm{m}}}{dt} - \frac{\omega(\omega + 2i\gamma)}{2(\gamma - i\omega)}\tilde{\mathbf{\Pi}}_{\mathrm{m}} = \frac{\epsilon_0\omega_{\mathrm{p}}^2}{2(\gamma - i\omega)}\tilde{\mathbf{E}}_{\mathrm{m}}, \tag{S.8}$$

which is Eq. (8) in the main article (where all tildas have been dropped by convention for readibility).

**2. Gain Medium**

The gain medium, made of emitters, can be modeled as a two-level system, where gain is achieved by introducing a phenomenological pump in addition to the typical thermal bath normally used to model purely absorbing elements. The two-level system is described through the optical Bloch equations in the density matrix formalism:

$$\frac{d\rho_{12}}{dt} - \left(i\omega_{12} - \frac{1}{\tau_2}\right)\rho_{12} = \frac{iN\boldsymbol{\mu}\cdot\mathbf{E}_{\mathrm{g}}}{\hbar} \tag{S.9}$$

$$\frac{dN}{dt} + \frac{N - \widetilde{N}}{\tau_1} = \frac{2i(\rho_{12} - \rho_{21})\boldsymbol{\mu}\cdot\mathbf{E}_{\mathrm{g}}}{\hbar}. \tag{S.10}$$

Here, the electric field of the gain medium, $\mathbf{E}_{\mathrm{g}}$, interacts with a single gain element of dipole moment $\boldsymbol{\mu}$. Also, $\rho_{ij}$ is the $i, j$ element of the density matrix. The time constants describing energy relaxation processes (spontaneous emission) and phase relaxation processes are, respectively, $\tilde{\tau}_1$ and $\tau_2$. We define the effective energy relaxation time $\tau_1$, which combines the effect of pumping and spontaneous emission on the population inversion $N$:

$$\tau_1 = \frac{\tilde{\tau}_1}{W\tilde{\tau}_1 + 1}.$$

The transition frequency between levels 1 and 2 (of respective energies $E_1$ and $E_2$) is

$$\omega_{\mathrm{g}} = \frac{E_2 - E_1}{\hbar}.$$

The quantity $N = \rho_{22} - \rho_{11}$ is the population inversion. When the gain element is subject to a phenomenological pump rate $W$, the corresponding equilibrium value of $N$ with the thermal reservoir is $N = \widetilde{N}$, given by

$$\widetilde{N} = \frac{W\tilde{\tau}_1 - 1}{W\tilde{\tau}_1 + 1}. \tag{S.11}$$

The presence of $\widetilde{N}$ in Equation S.10 means that, when the right-hand term of that equation is negligible, the population inversion is driven to $\tilde{N}$ in a time of the order of $\tau_1$. By choosing $\tilde{N} > 0$ here, we are effectively modeling a pump that drives the active elements to their excited state.

In this framework, the polarization of the gain medium, as arising from the collective behavior of the population of gain elements, can be calculated as the following integral:

$$\mathbf{P}_\mathrm{g} = \epsilon_0 \chi_\mathrm{b} \mathbf{E}_\mathrm{g} + \frac{n}{4\pi} \int\limits_{\Psi} [\rho_{12} + \rho_{12}^*] \boldsymbol{\mu} d\Psi \tag{S.12}$$

where $\chi_\mathrm{b}$ is the susceptibility of the dielectric host in which the gain elements are dispersed. The right side of expression S.12 reflects the contribution of a population of gain elements with volume density $n$ and dipole moments $\boldsymbol{\mu}$ activated by the element of the density matrix $\rho_{12}$ and its conjugate, which account for the probability of transition. The distribution of dipoles is assumed to be randomly oriented, so that the expression is averaged over all solid angles $\Psi$ through the integral. Expression S.12 shows that if the probability of transition were independent of the field in the gain region, the right term would just be averaged out. However, Equation S.9 has a driving term on the right-hand side that favors the transition of the gain elements whose dipole moment is parallel to the electric field $\mathbf{E}_\mathrm{g}$.

If we now define the active contribution to the polarization $\boldsymbol{\Pi}_\mathrm{g}$ as:

$$\boldsymbol{\Pi}_\mathrm{g} = \frac{n}{4\pi} \int\limits_{\Psi} [\rho_{12} + \rho_{12}^*] \boldsymbol{\mu} d\Psi, \tag{S.13}$$

expression S.12 can be rewritten as:

$$\mathbf{P}_\mathrm{g} = \epsilon_0 \chi_\mathrm{b} \mathbf{E}_\mathrm{g} + \boldsymbol{\Pi}_\mathrm{g}. \tag{S.14}$$

Also, considering that it is possible to demonstrate that

$$\int\limits_{\Psi} (\boldsymbol{\mu} \cdot \mathbf{E}_\mathrm{g}) \boldsymbol{\mu} d\Psi = \frac{4\pi}{3} \mu^2 \mathbf{E}_\mathrm{g},$$

one can rewrite the system of equations S.9-S.10 in terms of the time evolution of the dynamic part of the polarization in the gain medium:

$$\frac{d\boldsymbol{\Pi}_\mathrm{g}}{dt} - \left( i\omega_\mathrm{g} - \frac{1}{\tau_2} \right) \boldsymbol{\Pi}_\mathrm{g} = \frac{in\mu^2 N}{3\hbar} \mathbf{E}_\mathrm{g}, \tag{S.15}$$

$$\frac{dN}{dt} + \frac{N - \widetilde{N}}{\tau_1} = \frac{i}{n\hbar} (\boldsymbol{\Pi}_\mathrm{g} - \boldsymbol{\Pi}_\mathrm{g}^*) \cdot \mathbf{E}_\mathrm{g}. \tag{S.16}$$

Now, we use the rotating wave approximation again:

$$\mathbf{E}_\mathrm{g}(t) = \frac{1}{2} \left[ \tilde{\mathbf{E}}_\mathrm{g}(t) e^{-i\omega t} + \tilde{\mathbf{E}}_\mathrm{g}^*(t) e^{i\omega t} \right]$$

$$\boldsymbol{\Pi}_\mathrm{g}(t) = \frac{1}{2} [\tilde{\boldsymbol{\Pi}}_\mathrm{g}(t) e^{-i\omega t} + \tilde{\boldsymbol{\Pi}}_\mathrm{g}(t)^* e^{i\omega t}]$$

$$\mathbf{P}_\mathrm{g}(t) = \frac{1}{2} \left[ \tilde{\mathbf{P}}_\mathrm{g}(t) e^{-i\omega t} + \tilde{\mathbf{P}}_\mathrm{g}^*(t) e^{i\omega t} \right],$$

where $\tilde{\mathbf{E}}_h(t)$, $\tilde{\boldsymbol{\Pi}}_h(t)$ and $\tilde{\mathbf{P}}_h(t)$ represent again a slow dependency on time. When averaged over fast variations in time, (S.15) and (S.16) become:

$$\frac{d\tilde{\boldsymbol{\Pi}}_\mathrm{g}}{dt} - \left[ i(\omega - \omega_\mathrm{g}) - \frac{1}{\tau_2} \right] \tilde{\boldsymbol{\Pi}}_\mathrm{g} = \frac{in\mu^2 N}{3\hbar} \tilde{\mathbf{E}}_\mathrm{g}, \tag{S.17}$$

$$\frac{dN}{dt} + \frac{N - \widetilde{N}}{\tau_1} = -\frac{i}{2n\hbar} (\tilde{\boldsymbol{\Pi}}_\mathrm{g} \cdot \tilde{\mathbf{E}}_\mathrm{g}^* - \tilde{\boldsymbol{\Pi}}_\mathrm{g}^* \cdot \tilde{\mathbf{E}}_\mathrm{g}). \tag{S.18}$$

By defining the parameter $G$, which gives a measure of the level of gain brought into the system by the gain medium elements under pumping:

$$G = \frac{\tau_2 \mu^2}{3\hbar\epsilon_0} n\widetilde{N}, \tag{S.19}$$

one can rewrite the system of equations S.17-S.18 as:

$$\frac{d\tilde{\mathbf{\Pi}}_\mathrm{g}}{dt} - \left[i(\omega - \omega_\mathrm{g}) - \frac{1}{\tau_2}\right]\tilde{\mathbf{\Pi}}_\mathrm{g} = -\frac{i\epsilon_0 G}{\tau_2}\frac{N}{\widetilde{N}}\tilde{\mathbf{E}}_\mathrm{g}, \tag{S.20}$$

$$\frac{dN}{dt} + \frac{N - \widetilde{N}}{\tau_1} = -\frac{i}{2n\hbar}(\tilde{\mathbf{\Pi}}_\mathrm{g} \cdot \tilde{\mathbf{E}}_\mathrm{g}^* - \tilde{\mathbf{\Pi}}_\mathrm{g}^* \cdot \tilde{\mathbf{E}}_\mathrm{g}). \tag{S.21}$$

This system of equations governs the time evolution of the gain-enriched medium for different amounts of the gain quantity $G$. These equations are the same as Eqs. (6)–(7) in the main article (where it is reminded that all tildas were dropped out of notational convenience).

## 1.2 Steady-State Permittivities

From this point onwards, tildas will be meant implicitly for all fields and polarizations vectors and shall be removed, i.e., we are now exclusively dealing with the slowly-evolving, complex amplitudes introduced in the rotating-wave approximation.

When and if equation S.8 and system S.20-S.21 reach a steady state, one can calculate the permittivities $\epsilon_\mathrm{g}$ and $\epsilon_\mathrm{m}$ for the gain medium and the metal.

Starting with the metal permittivity, let us first consider the steady-state solution of equation S.8:

$$-\frac{\omega(\omega + 2i\gamma)}{2(\gamma - i\omega)}\mathbf{\Pi}_\mathrm{m} = \frac{\epsilon_0\omega_\mathrm{p}^2}{2(\gamma - i\omega)}\mathbf{E}_\mathrm{m},$$

from which one can calculate:

$$\mathbf{\Pi}_\mathrm{m} = -\frac{\epsilon_0\omega_\mathrm{p}^2}{\omega(\omega + 2i\gamma)}\mathbf{E}_\mathrm{m}.$$

Replacing the previous result in equation 2, one gets:

$$\mathbf{P}_\mathrm{m} = \epsilon_0\left[\chi_\infty - \frac{\omega_\mathrm{p}^2}{\omega(\omega + 2i\gamma)}\right]\mathbf{E}_\mathrm{m}.$$

Thus, the electric displacement is:

$$\mathbf{D}_\mathrm{m} = \epsilon_0\mathbf{E}_\mathrm{m} + \mathbf{P}_\mathrm{m}$$
$$\mathbf{D}_\mathrm{m} = \epsilon_0\left[1 + \chi_\infty - \frac{\omega_\mathrm{p}^2}{\omega(\omega + 2i\gamma)}\right]\mathbf{E}_\mathrm{m},$$

meaning that the metal steady state permittivity is:

$$\epsilon_\mathrm{m} = \epsilon_\infty - \frac{\epsilon_0\omega_\mathrm{p}^2}{\omega(\omega + 2i\gamma)}, \tag{S.22}$$

where $\epsilon_\infty = \epsilon_0(1 + \chi_\infty)$. Expression S.22 can be recognized as the Drude formula for metal permittivity, which appears as Eq. (10) in the main article.

Let us now switch to the gain medium. The steady-state solution of equation S.20 is:

$$-\left[i\left(\omega - \omega_\mathrm{g}\right) - \frac{1}{\tau_2}\right]\mathbf{\Pi}_\mathrm{g} = -\frac{i\epsilon_0 G N}{\tilde{N}\tau_2}\mathbf{E}_\mathrm{g},$$

from which one can calculate

$$\mathbf{\Pi}_{\rm g} = \frac{i\epsilon_0 GN}{\tilde{N}\tau_2}\frac{1}{i\left(\omega-\omega_{\rm g}\right)-\dfrac{1}{\tau_2}}\mathbf{E}_{\rm g} \tag{S.23}$$

$$= \frac{\epsilon_0 GN\Delta}{\tilde{N}}\frac{1}{2\left(\omega-\omega_{\rm g}\right)+i\Delta}\mathbf{E}_{\rm g}, \tag{S.24}$$

where we have defined the gain linewidth $\Delta = 2/\tau_2$.

Therefore, replacing this expression into the expression for the electric displacement in the gain medium, we get

$$\begin{aligned}\mathbf{D}_{\rm g} &= \epsilon_0\mathbf{E}_{\rm g} + \mathbf{P}_{\rm g}\\ &= \epsilon_{\rm b}\mathbf{E}_{\rm g} + \mathbf{\Pi}_{\rm g}\\ &= \left[\epsilon_{\rm b} + \frac{\epsilon_0 GN\Delta}{\tilde{N}}\frac{1}{2\left(\omega-\omega_{\rm g}\right)+i\Delta}\right]\mathbf{E}_{\rm g},\end{aligned}$$

where we define $\epsilon_{\rm b} = \epsilon_0(1+\chi_{\rm b})$, and the permittivity of the gain medium is:

$$\epsilon_{\rm g} = \epsilon_{\rm b} + \frac{\epsilon_0 G\Delta}{2\left(\omega-\omega_{\rm g}\right)+i\Delta}\frac{N}{\tilde{N}}. \tag{S.25}$$

To obtain the expression for $N$, we calculate the steady state solution of equation S.21, which is:

$$N = \tilde{N} - \frac{i\tau_1}{2n\hbar}\left(\mathbf{\Pi}_{\rm g}\cdot\mathbf{E}_{\rm g}^* - \mathbf{\Pi}_{\rm g}^*\cdot\mathbf{E}_{\rm g}\right). \tag{S.26}$$

By using equation S.24, we can calculate the right side of equation S.26, and obtain $N$:

$$\begin{aligned}N &= \tilde{N} - \frac{\epsilon_0 G\tau_1\Delta^2}{n\hbar\tilde{N}}N\frac{1}{\Delta^2+4\left(\omega-\omega_{\rm g}\right)^2}\left|\mathbf{E}_{\rm g}\right|^2\\ N&\left[\frac{\Delta^2+4\left(\omega-\omega_{\rm g}\right)^2+\dfrac{\epsilon_0 G\tau_1\Delta^2}{n\hbar\tilde{N}}\left|\mathbf{E}_{\rm g}\right|^2}{\Delta^2+4\left(\omega-\omega_{\rm g}\right)^2}\right] = \tilde{N}\\ N &= \tilde{N}\frac{\Delta^2+4\left(\omega-\omega_{\rm g}\right)^2}{\Delta^2+4\left(\omega-\omega_{\rm g}\right)^2+\dfrac{\epsilon_0 G\tau_1\Delta^2}{n\hbar\tilde{N}}\left|\mathbf{E}_{\rm g}\right|^2}.\end{aligned}$$

By introducing $E_{\rm sat} = \sqrt{n\hbar\tilde{N}/(\epsilon_0 G\tau_1)}$, which can be rewritten as $E_{\rm sat} = \hbar/\mu\sqrt{3/(\tau_1\tau_2)}$

$$N = \tilde{N}\frac{4\left(\omega-\omega_{\rm g}\right)^2+\Delta^2}{4\left(\omega-\omega_{\rm g}\right)^2+\Delta^2\left(1+\dfrac{\left|\mathbf{E}_{\rm g}\right|^2}{{E_{\rm sat}}^2}\right)}. \tag{S.27}$$

By replacing S.27 in equation S.24, we obtain:

$$\begin{aligned}\mathbf{\Pi}_{\rm g} &= \frac{\epsilon_0 G\Delta}{2\left(\omega-\omega_{\rm g}\right)+i\Delta}\frac{4\left(\omega-\omega_{\rm g}\right)^2+\Delta^2}{4\left(\omega-\omega_{\rm g}\right)^2+\Delta^2\left(1+\dfrac{\left|\mathbf{E}_{\rm g}\right|^2}{{E_{\rm sat}}^2}\right)}\mathbf{E}_{\rm g}\\ &= \epsilon_0 G\Delta\frac{2\left(\omega-\omega_{\rm g}\right)-i\Delta}{4\left(\omega-\omega_{\rm g}\right)^2+\Delta^2\left(1+\dfrac{\left|\mathbf{E}_{\rm g}\right|^2}{{E_{\rm sat}}^2}\right)}\mathbf{E}_{\rm g}.\end{aligned}$$

Now, we are able to calculate the electric displacement in the gain medium:

$$\mathbf{D}_{\rm g} = \epsilon_0\mathbf{E}_{\rm g} + \mathbf{P}_{\rm g} = \epsilon_0\left[1+\chi_{\rm b}+\frac{\left[2\left(\omega-\omega_{\rm g}\right)-i\Delta\right]G\Delta}{4\left(\omega-\omega_{\rm g}\right)^2+\Delta^2\left(1+\dfrac{\left|\mathbf{E}_{\rm g}\right|^2}{{E_{\rm sat}}^2}\right)}\right]\mathbf{E}_{\rm g},$$

from which we determine the permittivity, where $\epsilon_{\mathrm{b}} = \epsilon_0(1+\chi_{\mathrm{b}})$:

$$\epsilon_{\mathrm{g}} = \epsilon_{\mathrm{b}} + \frac{\epsilon_0 \left[2\left(\omega-\omega_{\mathrm{g}}\right) - i\Delta\right] G\Delta}{4\left(\omega-\omega_{\mathrm{g}}\right)^2 + \Delta^2 \left(1 + \dfrac{|\mathbf{E}_{\mathrm{g}}|^2}{{E_{\mathrm{sat}}}^2}\right)}. \tag{S.28}$$

which is the expression for permittivity given in Eq. (11) of the main text.

Equation S.28 is the permittivity of the gain media in the saturated case, i.e. when $N \neq \widetilde{N}$. On the other hand, in the "small-signal" regime, i.e. when $N = \tilde{N}$, we recover the linear Lorentzian permittivity:

$$\epsilon_{\mathrm{g}} = \epsilon_{\mathrm{b}} + \frac{\epsilon_0 G\Delta}{2(\omega-\omega_{\mathrm{g}}) + i\Delta}.$$

## 1.3 Boundary Conditions

The use of boundary conditions allows us to determine the coefficients of the Legendre polynomials present in the potentials defining the polarization and the electric field in the different regions of the system.

To proceed with the boundary conditions, we first calculate the radial and polar spherical coordinates components of the electric fields and polarizations, as derived from the potentials written down in equations (16) to (20) from the main article:

$$\begin{aligned}
E_{\mathrm{g}}^{r} &= -\frac{\partial\phi_1}{\partial r} = -p_0\cos\theta\\
E_{\mathrm{g}}^{\theta} &= -\frac{1}{r}\frac{\partial\phi_1}{\partial\theta} = p_0\sin\theta\\
\Pi_{\mathrm{g}}^{r} &= -\frac{\partial\psi_1}{\partial r} = -q_0\cos\theta\\
\Pi_{\mathrm{g}}^{\theta} &= -\frac{1}{r}\frac{\partial\psi_1}{\partial\theta} = q_0\sin\theta
\end{aligned}$$

$$\begin{aligned}
E_{\mathrm{m}}^{r} &= -\frac{\partial\phi_2}{\partial r} = -p_1\cos\theta + 2a^3\rho^3 p_2\frac{\cos\theta}{r^3}\\
E_{\mathrm{m}}^{\theta} &= -\frac{1}{r}\frac{\partial\phi_2}{\partial\theta} = p_1\sin\theta + a^3\rho^3 p_2\frac{\sin\theta}{r^3}\\
\Pi_{\mathrm{m}}^{r} &= -\frac{\partial\psi_2}{\partial r} = -q_1\cos\theta + 2a^3\rho^3 q_2\frac{\cos\theta}{r^3}\\
\Pi_{\mathrm{m}}^{\theta} &= -\frac{1}{r}\frac{\partial\psi_2}{\partial\theta} = q_1\sin\theta + a^3\rho^3 q_2\frac{\sin\theta}{r^3}
\end{aligned}$$

$$\begin{aligned}
E_{\mathrm{e}}^{r} &= -\frac{\partial\phi_3}{\partial r} = E_0\cos\theta + 2a^3 p_3\frac{\cos\theta}{r^3}\\
E_{\mathrm{e}}^{\theta} &= -\frac{1}{r}\frac{\partial\phi_3}{\partial\theta} = -E_0\sin\theta + a^3 p_3\frac{\sin\theta}{r^3}.
\end{aligned}$$

**1. Metal Outer boundary $r = a$:**

• Radial continuity:

$$\begin{aligned}
&D_{\mathrm{m}}^{r}|_{r=a} = D_{\mathrm{e}}^{r}|_{r=a}\\
&\left(\epsilon_0 E_{\mathrm{m}}^{r} + P_{\mathrm{m}}^{r}\right)|_{r=a} = \left(\epsilon_0 E_{\mathrm{e}}^{r} + P_{\mathrm{e}}^{r}\right)|_{r=a}\\
&\epsilon_\infty E_{\mathrm{m}}^{r}|_{r=a} + \Pi_{\mathrm{m}}^{r}|_{r=a} = \epsilon_{\mathrm{e}} E_{\mathrm{e}}^{r}|_{r=a}\\
&\epsilon_\infty\left(-p_1\cos\theta + 2\rho^3 p_2\cos\theta\right) - q_1\cos\theta + 2\rho^3 q_2\cos\theta = \epsilon_{\mathrm{e}}\left(E_0\cos\theta + 2p_3\cos\theta\right)\\
&-\epsilon_\infty p_1 + 2\epsilon_\infty\rho^3 p_2 - q_1 + 2\rho^3 q_2 = \epsilon_{\mathrm{e}} E_0 + 2\epsilon_{\mathrm{e}} p_3
\end{aligned}$$

• Tangential continuity:

$$
\begin{aligned}
&E^{\theta}_{\mathrm{m}}|_{r=a} = E^{\theta}_{\mathrm{e}}|_{r=a} \\
&p_1 \sin\theta + \rho^3 \sin\theta p_2 = -E_0 \sin\theta + \sin\theta p_3 \\
&p_1 + \rho^3 p_2 = -E_0 + p_3
\end{aligned}
$$

**2. Gain-metal boundary at $r = \rho a$:** • Radial continuity:

$$
\begin{aligned}
&D^{r}_{\mathrm{m}}|_{r=\rho a} = D^{r}_{\mathrm{g}}|_{r=\rho a} \\
&\left(\epsilon_0 E^r_{\mathrm{m}} + P^r_{\mathrm{m}}\right)|_{r=\rho a} = \left(\epsilon_0 E^r_{\mathrm{g}} + P^r_{\mathrm{g}}\right)|_{r=\rho a} \\
&\epsilon_\infty E^r_{\mathrm{m}}|_{r=\rho a} + \Pi^r_{\mathrm{m}}|_{r=\rho a} = \epsilon_{\mathrm{b}} E^r_{\mathrm{g}}|_{r=\rho a} + \Pi^r_{\mathrm{g}}|_{r=\rho a} \\
&\epsilon_\infty \left(-p_1 \cos\theta + 2\rho^3 a^3 p_2 \frac{\cos\theta}{\rho^3 a^3}\right) - q_1 \cos\theta + 2\rho^3 a^3 q_2 \frac{\cos\theta}{\rho^3 a^3} = -\epsilon_{\mathrm{b}} p_0 \cos\theta - q_0 \cos\theta \\
&-\epsilon_\infty p_1 + 2\epsilon_\infty p_2 - q_1 + 2q_2 = -\epsilon_{\mathrm{b}} p_0 - q_0
\end{aligned}
$$

• Tangential continuity:

$$
\begin{aligned}
&E^{\theta}_{\mathrm{m}}|_{r=\rho a} = E^{\theta}_{\mathrm{g}}|_{r=\rho a} \\
&p_1 \sin\theta + \frac{\rho^3 a^3 \sin\theta}{\rho^3 a^3} p_2 = p_0 \sin\theta \\
&p_1 + p_2 = p_0
\end{aligned}
$$

Therefore, from the boundary conditions, we obtain:

$$p_3 = \frac{-\epsilon_\infty p_1 + 2\epsilon_\infty \rho^3 p_2 - q_1 + 2\rho^3 q_2 - \epsilon_{\mathrm{e}} E_0}{2\epsilon_{\mathrm{e}}} \tag{S.29}$$

$$p_2 = \frac{(\epsilon_{\mathrm{b}} - \epsilon_\infty)(p_3 - E_0) + q_0 - q_1 + 2q_2}{-2\epsilon_\infty - \epsilon_{\mathrm{b}} + \rho^3 (\epsilon_{\mathrm{b}} - \epsilon_\infty)} \tag{S.30}$$

$$p_1 = p_3 - \rho^3 p_2 - E_0 \tag{S.31}$$

$$p_0 = p_1 + p_2. \tag{S.32}$$

## 1.4 Steady-State Polarizability $\alpha$

Let us now prove that Eq. (34) in the main article holds, with the classical expression for the polarizability $\alpha$ as written in Eq. (28) :

$$\frac{\alpha}{4\pi a^3} = \frac{(\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{e}})(\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}}) + \rho^3(\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{m}})(\epsilon_{\mathrm{e}} + 2\epsilon_{\mathrm{m}})}{(\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}})(\epsilon_{\mathrm{m}} + 2\epsilon_{\mathrm{e}}) + 2\rho^3(\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{m}})(\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{e}})} \tag{S.33}$$

To demonstrate this statement, we begin with calculating the steady-state solutions of equations (48) to (50) of the main article:

$$q_0 = -\frac{\Gamma_{\mathrm{g}} N}{\Omega_{\mathrm{g}}} p_0 \tag{S.34}$$

$$q_1 = -\frac{\Gamma_{\mathrm{m}}}{\Omega_{\mathrm{m}}} p_1 \tag{S.35}$$

$$q_2 = -\frac{\Gamma_{\mathrm{m}}}{\Omega_{\mathrm{m}}} p_2. \tag{S.36}$$

From equation (52) of the article, we can deduce that

$$-2i\Omega_{\mathrm{g}} = 2(\omega - \omega_{\mathrm{g}}) + i\Delta.$$

Recall now the expression for the permittivity of the gain medium in equation S.25:

$$\epsilon_{\mathrm{g}} = \epsilon_{\mathrm{b}} + \frac{\epsilon_0 G N \Delta}{\tilde{N}} \frac{1}{2(\omega - \omega_{\mathrm{g}}) + i\Delta}. \tag{S.37}$$

Replacing the expression for $-2i\Omega_{\mathrm{g}}$ in equation S.37, we obtain that

$$\epsilon_{\mathrm{g}} = \epsilon_{\mathrm{b}} - \frac{\epsilon_0 G N \Delta}{\tilde{N}} \frac{1}{2i\Omega_{\mathrm{g}}}. \tag{S.38}$$

Also, according to equation (53) in the article:

$$G = \frac{2i\tilde{N}}{\epsilon_0 \Delta} \Gamma_{\mathrm{g}},$$

thus, equation (S.25) becomes

$$\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{b}} = -\frac{N\Gamma_{\mathrm{g}}}{\Omega_{\mathrm{g}}}.$$

On the other hand, from equations (54) and (55), we obtain that

$$\begin{aligned} \frac{\Gamma_{\mathrm{m}}}{\Omega_{\mathrm{m}}} &= \frac{\epsilon_0 \omega_{\mathrm{p}}^2}{\omega(\omega + 2i\gamma)} \\ &= \epsilon_\infty - \epsilon_{\mathrm{m}}. \end{aligned}$$

Consequently, $q_0$, $q_1$, and $q_2$ can be written as

$$\begin{aligned} q_0 &= (\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{b}})\, p_0 \\ q_1 &= (\epsilon_{\mathrm{m}} - \epsilon_\infty)\, p_1 \\ q_2 &= (\epsilon_{\mathrm{m}} - \epsilon_\infty)\, p_2. \end{aligned} \tag{S.39}$$

We now simplify equation (44) of the main article by substituting the set of equations S.39 into it:

$$p_3 = \frac{-\epsilon_\infty p_1 + 2\epsilon_\infty \rho^3 p_2 - (\epsilon_{\mathrm{m}} - \epsilon_\infty)\, p_1 + 2\rho^3 (\epsilon_{\mathrm{m}} - \epsilon_\infty)\, p_2 - \epsilon_{\mathrm{e}} E_0}{2\epsilon_{\mathrm{e}}} \tag{S.40}$$

$$= \frac{-\epsilon_{\mathrm{m}} p_1 + 2\rho^3 \epsilon_{\mathrm{m}} p_2 - \epsilon_{\mathrm{e}} E_0}{2\epsilon_{\mathrm{e}}}. \tag{S.41}$$

Next, we simplify equation (45) by using equation (46) and substituting with the set S.39 again:

$$\begin{aligned} p_2 &= \frac{(\epsilon_{\mathrm{b}} - \epsilon_\infty)(p_3 - E_0) + (\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{b}})\, p_0 - (\epsilon_{\mathrm{m}} - \epsilon_\infty)\, p_1 + 2(\epsilon_{\mathrm{m}} - \epsilon_\infty)\, p_2}{-2\epsilon_\infty - \epsilon_{\mathrm{b}} + \rho^3 (\epsilon_{\mathrm{b}} - \epsilon_\infty)} \\ &= \frac{(\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{b}} - \epsilon_{\mathrm{m}} + \epsilon_\infty)\left(p_3 - \rho^3 p_2 - E_0\right) + (\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{b}} + 2\epsilon_{\mathrm{m}} - 2\epsilon_\infty)\, p_2}{-2\epsilon_\infty - \epsilon_{\mathrm{b}} + \rho^3 (\epsilon_{\mathrm{b}} - \epsilon_\infty)} \\ &= \frac{(\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{g}})(p_3 - E_0)}{\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}} + \rho^3 (\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{g}})}. \end{aligned}$$

We replace this result in equation 46 to obtain $p_1$ in terms of $p_3$ and $E_0$:

$$\begin{aligned} p_1 &= -E_0 - \frac{\rho^3 (\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{g}})(p_3 - E_0)}{\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}} + \rho^3 (\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{g}})} + p_3 \\ &= \frac{(\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}})(p_3 - E_0)}{\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}} + \rho^3 (\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{g}})}. \end{aligned}$$

By using these expressions of $p_2$ and $p_1$ in equation S.41, we obtain:

$$p_3 = -\frac{\epsilon_{\mathrm{m}}}{2\epsilon_{\mathrm{e}}} \frac{(\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}})(p_3 - E_0)}{\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}} + \rho^3 (\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{g}})} + \frac{\rho^3 \epsilon_{\mathrm{m}}}{\epsilon_{\mathrm{e}}} \frac{(\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{g}})(p_3 - E_0)}{\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}} + \rho^3 (\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{g}})} - \frac{E_0}{2},$$

which after rearrangement, gives the proportionality relation between $p_3$ and $E_0$:

$$p_3 = \frac{(\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}})(\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{e}}) + \rho^3 (\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{m}})(\epsilon_{\mathrm{e}} + 2\epsilon_{\mathrm{m}})}{(\epsilon_{\mathrm{g}} + 2\epsilon_{\mathrm{m}})(2\epsilon_{\mathrm{e}} + \epsilon_{\mathrm{m}}) + 2\rho^3 (\epsilon_{\mathrm{g}} - \epsilon_{\mathrm{m}})(\epsilon_{\mathrm{m}} - \epsilon_{\mathrm{e}})} E_0. \tag{S.42}$$

Since by definition of the polarizability [see Eqs. (21) and (29) of the article], we have $p_3 = \alpha E_0/(4\pi a^3)$, we deduce that $\alpha$ has indeed the form of equation S.33, or Eq. (28) in the main text.

## 1.5 The Geometry Matrix

To express the matrix system of equations as stated in (60) of the article, let us recall expressions (44)–(47):

$$
\begin{aligned}
p_3 =& \frac{-\epsilon_\infty p_1 + 2\epsilon_\infty \rho^3 p_2 - q_1 + 2\rho^3 q_2 - \epsilon_{\mathrm{e}} E_0}{2\epsilon_{\mathrm{e}}} \\
p_2 =& \frac{(\epsilon_{\mathrm{b}} - \epsilon_\infty)(p_3 - E_0) + q_0 - q_1 + 2q_2}{-2\epsilon_\infty - \epsilon_{\mathrm{b}} + \rho^3(\epsilon_{\mathrm{b}} - \epsilon_\infty)}; \\
p_1 =& p_3 - \rho^3 p_2 - E_0; \\
p_0 =& p_1 + p_2.
\end{aligned}
$$

It is now necessary to write all of these relations as linear functions of the main variables $q_0$, $q_1$, $q_2$, as well as $E_0$.

By replacing equations (46) and (45) into equation (44), we get:

$$
p_3 = \frac{\epsilon_\infty}{2\epsilon_{\mathrm{e}}}\left(E_0 + \rho^3 p_2 - p_3\right) + \frac{2\rho^3\epsilon_\infty}{2\epsilon_{\mathrm{e}}}\frac{(\epsilon_{\mathrm{b}} - \epsilon_\infty)(p_3 - E_0) + q_0 - q_1 + 2q_2}{-2\epsilon_\infty - \epsilon_{\mathrm{b}} + \rho^3(\epsilon_{\mathrm{b}} - \epsilon_\infty)} \tag{S.43}
$$

$$
+ \frac{-q_1 + 2\rho^3 q_2 - \epsilon_{\mathrm{e}} E_0}{2\epsilon_{\mathrm{e}}} \tag{S.44}
$$

$$
= -\frac{3\rho^3\epsilon_\infty}{D} q_0 - \frac{(1-\rho^3)(\epsilon_{\mathrm{b}} + 2\epsilon_\infty)}{D} q_1 + \frac{2\rho^3(1-\rho^3)(\epsilon_{\mathrm{b}} - \epsilon_\infty)}{D} q_2 \tag{S.45}
$$

$$
+ \frac{(\epsilon_\infty - \epsilon_{\mathrm{e}})(\epsilon_{\mathrm{b}} + 2\epsilon_\infty) + \rho^3(\epsilon_{\mathrm{b}} - \epsilon_\infty)(\epsilon_{\mathrm{e}} + 2\epsilon_\infty)}{D} E_0, \tag{S.46}
$$

where, in order to have more compact formulas, we define:

$$
D = (\epsilon_\infty + 2\epsilon_{\mathrm{e}})(\epsilon_{\mathrm{b}} + 2\epsilon_\infty) + 2\rho^3(\epsilon_{\mathrm{b}} - \epsilon_\infty)(\epsilon_\infty - \epsilon_{\mathrm{e}}).
$$

We then replace equation S.46 into equation (45), and obtain:

$$
p_2 = -\frac{\epsilon_\infty + 2\epsilon_{\mathrm{e}}}{D} q_0 + \frac{\epsilon_{\mathrm{b}} + 2\epsilon_{\mathrm{e}}}{D} q_1 - \frac{2[(\epsilon_\infty + 2\epsilon_2) + \rho^3(\epsilon_{\mathrm{b}} - \epsilon_\infty)]}{D} q_2 + \frac{3\epsilon_2(\epsilon_{\mathrm{b}} - \epsilon_\infty)}{D} E_0. \tag{S.47}
$$

We can now calculate $p_1$ by replacing S.46 and S.47 into (46):

$$
\begin{aligned}
p_1 = \frac{2\rho^3(\epsilon_{\mathrm{e}} - \epsilon_\infty)}{D} q_0 - & \frac{\rho^3(\epsilon_{\mathrm{b}} + 2\epsilon_{\mathrm{e}}) + (1-\rho^3)(\epsilon_{\mathrm{b}} + 2\epsilon_\infty)}{D} q_1 + \\
& + \frac{2\rho^3(\epsilon_{\mathrm{b}} + 2\epsilon_{\mathrm{e}})}{D} q_2 - \frac{3\epsilon_{\mathrm{e}}(\epsilon_{\mathrm{b}} + 2\epsilon_\infty)}{D} E_0.
\end{aligned} \tag{S.48}
$$

Finally, we calculate $p_0$:

$$
\begin{aligned}
p_0 = -\frac{(\epsilon_\infty + 2\epsilon_{\mathrm{e}}) + 2\rho^3(\epsilon_\infty - \epsilon_{\mathrm{e}})}{D} q_0 - & \frac{2(1-\rho^3)(\epsilon_\infty - \epsilon_{\mathrm{e}})}{D} q_1 + \\
- & \frac{2(1-\rho^3)(\epsilon_\infty + 2\epsilon_{\mathrm{e}})}{D} q_2 - \frac{9\epsilon_{\mathrm{e}}\epsilon_\infty}{D} E_0.
\end{aligned} \tag{S.49}
$$

We can thus rewrite the obtained expressions S.46–S.49 for $p_3$, $p_2$, $p_1$, and $p_0$ in the following form:

$$
\begin{aligned}
p_0 =& p_{00} q_0 + p_{01} q_1 + p_{02} q_2 + p_{03} E_0 \\
p_1 =& p_{10} q_0 + p_{11} q_1 + p_{12} q_2 + p_{13} E_0 \\
p_2 =& p_{20} q_0 + p_{21} q_1 + p_{22} q_2 + p_{23} E_0 \\
p_3 =& p_{30} q_0 + p_{31} q_1 + p_{32} q_2 + p_{33} E_0.
\end{aligned}
$$

where the $p_{ij}$ coefficients have been defined as:

$$p_{00} = -\frac{\epsilon_\infty + 2\epsilon_\mathrm{e} + 2\rho^3(\epsilon_\infty - \epsilon_\mathrm{e})}{D}$$
$$p_{01} = -\frac{2(1-\rho^3)(\epsilon_\infty - \epsilon_\mathrm{e})}{D}$$
$$p_{02} = -\frac{2(1-\rho^3)(\epsilon_\infty + 2\epsilon_\mathrm{e})}{D}$$
$$p_{03} = -\frac{9\epsilon_\mathrm{e}\epsilon_\infty}{D}$$
$$p_{10} = \frac{2\rho^3(\epsilon_\mathrm{e} - \epsilon_\infty)}{D}$$
$$p_{11} = -\frac{\rho^3(\epsilon_\mathrm{b} + 2\epsilon_\mathrm{e}) + (1-\rho^3)(\epsilon_\mathrm{b} + 2\epsilon_\infty)}{D}$$
$$p_{12} = \frac{2\rho^3(\epsilon_\mathrm{b} + 2\epsilon_\mathrm{e})}{D}$$
$$p_{13} = -\frac{3\epsilon_\mathrm{e}(\epsilon_\mathrm{b} + 2\epsilon_\infty)}{D}$$
$$p_{20} = -\frac{\epsilon_\infty + 2\epsilon_\mathrm{e}}{D}$$
$$p_{21} = \frac{\epsilon_\mathrm{b} + 2\epsilon_\mathrm{e}}{D}$$
$$p_{22} = -\frac{2[\epsilon_\infty + 2\epsilon_\mathrm{e} + \rho^3(\epsilon_\mathrm{b} - \epsilon_\infty)]}{D}$$
$$p_{23} = \frac{3\epsilon_\mathrm{e}(\epsilon_\mathrm{b} - \epsilon_\infty)}{D}$$
$$p_{30} = -\frac{3\rho^3\epsilon_\infty}{D}$$

$$p_{31} = -\frac{(1-\rho^3)(\epsilon_\mathrm{b} + 2\epsilon_\infty)}{D}$$
$$p_{32} = \frac{2\rho^3(1-\rho^3)(\epsilon_\mathrm{b} - \epsilon_\infty)}{D}$$
$$p_{33} = \frac{(\epsilon_\infty - \epsilon_\mathrm{e})(\epsilon_\mathrm{b} + 2\epsilon_\infty) + \rho^3(\epsilon_\mathrm{b} - \epsilon_\infty)(\epsilon_\mathrm{e} + 2\epsilon_\infty)}{D}.$$

This gives us the system of Eqs. (56)-(59) as written down in the main article.

# 2 Exciting field and Emission Intensity

We here give some indication on how the dynamics of the nanoshell in the lasing regime was obtained, as exposed in the “Above threshold” section of the main article.

Since we were interested in investigating situations of free lasing (no external drive), it seemed physical to use zero-field initial conditions, and zero external probe, and then leave the lasing instability to grow out of the numerical noise. This procedure, however, gives rise to prohibitively long computational times, and becomes especially inefficient when computing spectra including many frequency points. This is why as a numerical trick, we in fact applied a minute probe field $E_0$ acting like a “seed” and driving the initial steps of the instability faster. To produce the figures shown in Section 6, we chose to apply a field value $E_0 = 10^{-8}E_\mathrm{sat}$.

To make sure nonetheless that the results we obtained were in the free lasing regime and that the presence of the small $E_0$ did not generate any forced oscillation regime, we verified that the final results did not depend on the value chosen for $E_0$. This is illustrated in the following Figure, where the emitted intensity $I_{\mathrm{em}}(t)$ has been calculated in the same conditions as Figs. 6 to 8 of the main article, namely, at the frequency $\hbar\omega = 2.811$ eV and with a gain level $G = 1.01\,G_{\mathrm{th}}$. Results are shown for $E_0 = 10^{-10}$ to $10^{-7}E_{\mathrm{sat}}$: it is seen that the obtained responses are indeed all exactly the same to within some time translation, corresponding to the onset time of the lasing instability.

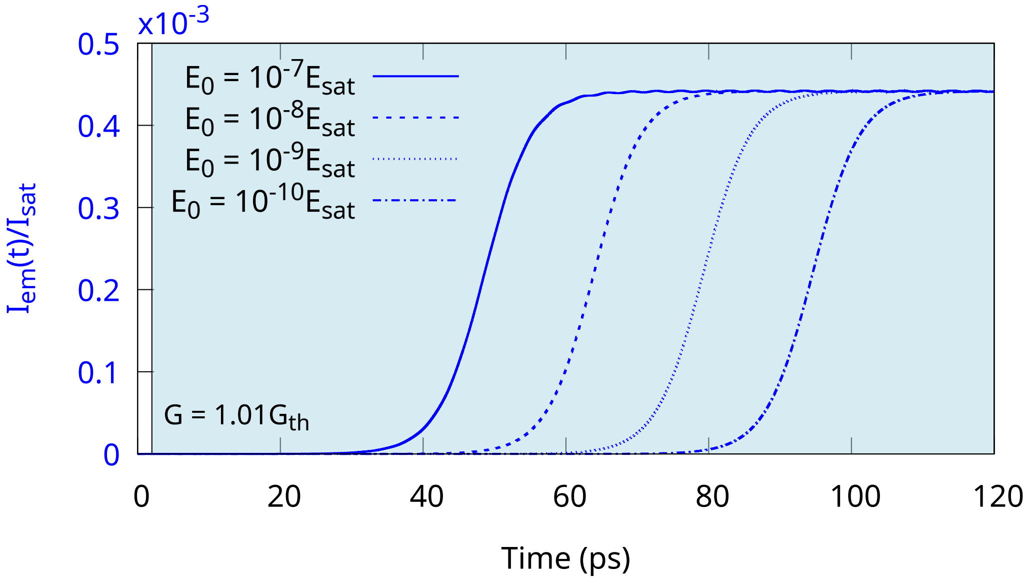


**Fig. 1:** Identical results (to within a time translation) were obtained for all values of the field $E_0$ used to accelerate the numerical onset of the lasing instability, confirming that the nanolaser is in a free lasing regime.