S1. THE EXCITATION ENHANCEMENT AND THE QUANTUM YIELD

FIG. S1. The dependence of the excitation enhancement on the gap size $g$ and the wavelength $\lambda$ as computed from LRA, TF-HT and QHT for $R = 2.5; 5; 10$ nm.

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FIG. S2. The dependence of the quantum yield on the gap size $g$ and the wavelength $\lambda$ as computed from LRA, TF-HT and QHT for $R = 2.5; 5; 10$ nm.
S2. THE COMPUTATION DOMAIN AND THE MESHING.

FIG. S3. The schematic images for the computation domain - (a), the meshing style used - (b) - (d).
FIG. S4. Current density as obtained from LRA, TF and QHT at $\lambda = 400$ nm for $g = 0.1$ nm and $R = 10$ nm.
S4. THE POTENTIAL

The first-order terms in the potential

$$\left( \frac{\delta G[n]}{\delta n} \right)_1 = \left( \frac{\delta E_{\text{XC}}^{\text{LDA}}}{\delta n} \right)_1 + \left( \frac{\delta T_s}{\delta n} \right)_1$$

are presented below.

For the exchange-correlation potential, it is given by:

$$\left( \frac{\delta E_{\text{XC}}^{\text{LDA}}}{\delta n} \right)_1 = (E_h) \left( -a_0 \frac{4}{9} c_X n_0^{-2/3} n_1 + a_0^3 \mu'_C[n_0] n_1 \right),$$

where

$$\mu'_C[n] = \begin{cases} -\frac{4\pi}{27} \left[ \left( 1 + 2 \ln(r_s) \right) c r_s + \frac{2}{3} d r_s^3 \right] r_s^2, & r_s < 1, \\ \frac{5\beta_1 \sqrt{r_s} (7\beta_2^2 + 8\beta_2 r_s + 21\beta_1 \beta_2 r_s^2) + 16\beta_3^2 r_s^2}{(1 + \beta_1 \sqrt{r_s} + \beta_2 r_s)^3}, & r_s \geq 1, \end{cases}$$

with $c_X = \frac{3}{4} \left( \frac{2}{3} \right)^{1/3}$ and the values of the coefficients are $a = 0.0311$, $b = -0.048$, $c = 0.002$, $d = -0.0116$, $\alpha = -0.1423$, $\beta_1 = 1.0529$, and $\beta_2 = 0.3334$.

The expression for $\left( \frac{\delta T_s}{\delta n} \right)_1$ is:

$$\left( \frac{\delta T_s}{\delta n} \right)_1 = \left( \frac{\delta T_s^I}{\delta n} \right)_1 + \left( \frac{\delta T_s^{II}}{\delta n} \right)_1 + \left( \frac{\delta T_s^{III}}{\delta n} \right)_1,$$

where

$$\left( \frac{\delta T_s^I}{\delta n} \right)_1 = \tau_n^{(0)} n_1$$

$$\left( \frac{\delta T_s^{II}}{\delta n} \right)_1 = -2 \tau^{(0)}_{nnw} |\nabla n_0|^2 n_1 - 2 \tau^{(0)}_{nww} \left[ n_1 \nabla^2 n_0 + \nabla n_0 \cdot \nabla n_1 \right] - 2 \tau^{(0)}_{n} \nabla^2 n_1$$

$$- 2 \tau^{(0)}_{nww} \left[ 2 (\nabla n_0 \cdot \nabla n_1) \nabla^2 n_0 + \nabla \left( |\nabla n_0|^2 \right) \cdot \nabla n_1 + 2 \nabla n_0 \cdot \nabla (\nabla n_0 \cdot \nabla n_1) \right]$$

$$- 4 \tau^{(0)}_{nww} (\nabla n_0 \cdot \nabla n_1) \nabla n_0 \cdot \nabla \left( |\nabla n_0|^2 \right)$$

$$- 2 \tau^{(0)}_{nww} \left[ 2 (\nabla n_0 \cdot \nabla n_1) |\nabla n_0|^2 + \nabla n_0 \cdot \nabla \left( |\nabla n_0|^2 \right) n_1 \right],$$

$$\left( \frac{\delta T_s^{III}}{\delta n} \right)_1 = \tau^{(0)}_{nnnq} |\nabla n_0|^2 n_1 + \tau^{(0)}_{nnq} \left[ 2 \nabla n_0 \cdot \nabla n_1 + n_1 \nabla^2 n_0 \right] + 2 \tau^{(0)}_{nq} \nabla^2 n_1$$

$$+ \tau^{(0)}_{nnnq} \left[ |\nabla n_0|^2 \nabla^2 n_1 + 2 \nabla n_0 \cdot \nabla (\nabla^2 n_0) \right]$$

$$+ \tau^{(0)}_{nnq} \left[ \nabla^2 n_0 \nabla^2 n_1 + 2 \nabla n_0 \cdot \nabla (\nabla^2 n_1) + 2 \nabla n_0 \cdot \nabla (\nabla^2 n_1) + \nabla^2 (\nabla^2 n_0) n_1 \right]$$

$$+ \tau^{(0)}_{nq} \nabla^2 (\nabla^2 n_1)$$

$$+ \tau^{(0)}_{nqq} \left[ 2 (\nabla (\nabla^2 n_0) \cdot \nabla (\nabla^2 n_1)) + \nabla^2 (\nabla^2 n_0) \nabla^2 n_1 \right]$$

$$+ \tau^{(0)}_{nqq} \left[ 2 (\nabla n_0 \cdot \nabla (\nabla^2 n_0)) \nabla^2 n_1 + \nabla (\nabla^2 n_0)^2 n_1 \right]$$

$$+ \tau^{(0)}_{qq} \nabla (\nabla^2 n_0)^2 \nabla^2 n_1.$$
The subscripts \( i = n, w, q \) denote the partial derivatives, and the superscript \((0)\) means that the function is evaluated at \( n = n_0 \).

The potentials for each scenario considered in the article are as follows.

- For LRA there is no functional and hence \((\frac{\delta G[n]}{\delta n})_1 = 0\).
- For TF-HT \((\frac{\delta G[n]}{\delta n})_1\) is given by the Eq. \((S5a)\).
- For QHT, Eqs. \((S5a)\) and \((S5b)\) are considered.
- For QHT-PGSL we use Eqs. \((S5a)-(S5i)\).
- And finally, for QHT-PGSLN all the equations in \((S5)\) have to be accounted for.