Electronic Supplementary Material (ESI) for Nanoscale.

Electronic Supplementary Information

Excitation of plasmonic nanoantennas with nonresonant and resonant electron tunneling

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SI-1. SPP mode in nanowire

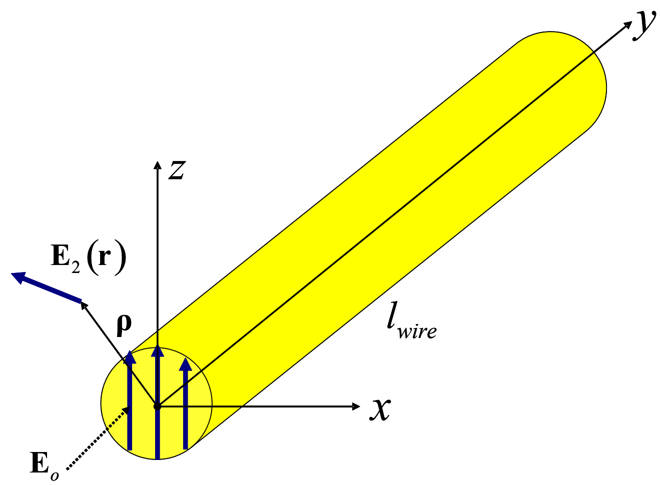


Fig. 1-SI.

Figure 1-SI shows a metal nanowire of the length l_{wire} and of radius a ($a << l_{wire}$), embedded into dielectric medium with the permittivity ε_d. The permittivity of the metal is $\varepsilon_m(\omega) = 1 - \omega_{pl}^2 / \omega^2$ In our description, we are following the arguments of [Novotny L. and Hecht B., *Principles of Nano-Optics*, Chapter 12 - Surface Plasmons, 12.3.1 Plasmons supported by wires and particles]. The fundamental surface plasmon mode in the quasistatic approximation has the frequency

$$\omega_{sp} = \omega_{pl} \Big/ \sqrt{1+\varepsilon_d} \qquad \text{(SI-1)}$$

The electric field *inside* the nanowire is homogeneous and is assumed to be along the *z* axis:

$$\mathbf{E}_1(\mathbf{r},t) = \mathbf{E}_o \cdot e^{-i\omega_{sp}t} + c.c. \qquad \text{(SI-2)}$$

Outside the nanowire the field $\mathbf{E}_2$ is inhomogeneous: $\mathbf{E}_2(\mathbf{r},t) = \mathbf{E}_2(\mathbf{r}) \cdot e^{-i\omega_{sp}t} + c.c$. One can show that outside the nanowire

$$\left|\mathbf{E}_2(\mathbf{r})\right|^2 = \left|E_o\right|^2 \frac{a^4}{\rho^4}, \qquad \rho > a \tag{SI-3}$$

where ρ is the distance from the axis of nanowire. The time-averaged energy of the field is given by [Landau, L. D.; Lifshitz, E. M. *Electrodynamics of Continuous Media, Second Edition*; Elsevier Science Ltd., 1984]:

$$\bar{w} = \varepsilon_o \frac{d\left[\omega\varepsilon_m(\omega)\right]}{d\omega}\left|\mathbf{E}_o\right|^2 + \mu_o\mu\left|\mathbf{H}\right|^2 \qquad \text{(inside nanowire, } \rho < a\text{)} \tag{SI-4}$$

$$\bar{w} = \varepsilon_o\varepsilon_d \cdot \left|\mathbf{E}_2(\mathbf{r})\right|^2 + \mu_o\mu\left|\mathbf{H}\right|^2 \qquad \text{(outside nanowire, } \rho > a\text{)} \tag{SI-5}$$

Below, we use Eqs. (SI-4)-(SI-5) for the expression of the quasistatic mode, dropping the term $\mu_o\mu\left|\mathbf{H}\right|^2$. The energy W_{mode} in the mode is given by spatial integration (see Fig.1-SI):

$$W_{\text{mode}} = \int dyd\boldsymbol{\rho}\bar{U} = l_{wire} \cdot \varepsilon_o\left|E_o\right|^2 \pi a^2 \left(\left.\frac{d(\omega\varepsilon_m)}{d\omega}\right|_{\omega=\omega_{sp}} + \varepsilon_d \right) \tag{SI-6}$$

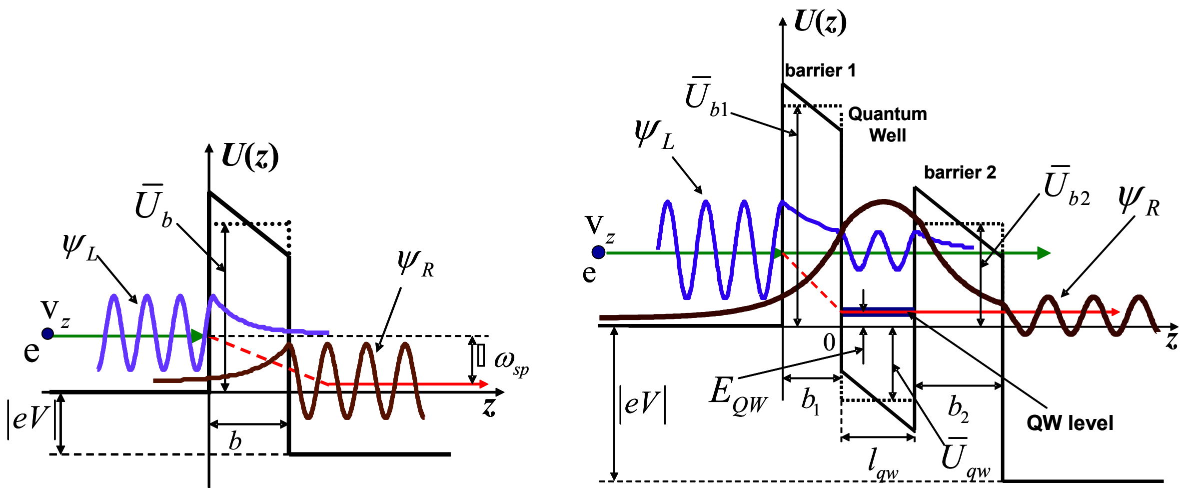
By taking into account Eq. (1), we have finally

$$W_{\text{mode}} = 2l_{wire} \cdot \pi a^2 \varepsilon_o \left(1+\varepsilon_d\right)\left|E_o\right|^2 \tag{SI-7}$$

One should note at the resonant frequency $\omega_{sp} = \omega_{pl}\big/\sqrt{1+\varepsilon_d}$, the permittivity of nanoantenna metal $\varepsilon_{metal} = -\varepsilon_d$. This implies that if we insert *very thin* dielectric layer inside the nanoantenna normally to the homogeneous field (see Fig.1-SI), then the field inside the dielectric will be the same *in magnitude* as in the nanoantenna. If we further insert *metallic* layer inside the dielectric layer, then the field inside this metallic layer will coincides with the field in nanoantenna.

This motivates usage of above formulas for the fields, obtained for pure metallic nanoantenna (see Fig.1-SI), in calculations of tunneling in nanoantennas with very thin dielectric and metallic layers inside the nanoantenna as we are doing in the paper.

SI-2. Wave functions $\psi_L(z)$ and $\psi_R(z)$ for single barrier, nonresonant tunneling structures



(a) (b)

Fig. 2-SI. (a) Potential barrier $U(z)$ for tunneling electron in the case of a single dielectric layer between two metal electrodes with a schematic illustration of the wave functions $\psi_L(z)$ and $\psi_R(z)$ of an electron in the left and in the right electrodes, respectively. The tails of these wave functions penetrate and overlap into the barrier. (b) Potential barrier $U(z)$ for a double-barrier structure with a Quantum Well located between two barriers, together with a schematic illustration of the wave functions $\psi_L(z)$ and $\psi_R(z)$ of electron in the left and right metals. The energy level of the QW is shown as dark blue. The green arrow is the elastic tunneling, the red arrow is the inelastic tunneling with emission of plasmon and the dotted lines show the averaged potentials insides the barriers and the QW.

The potential energy $U(z)$ for electron in tunneling structure with single barrier is shown in Fig. 2a-SI. In calculations of $\psi_L(z)$ and $\psi_R(z)$, we are using the following approximations:

(1) Generally speaking, $U(z)$ changes inside the barrier, in particular, due to the built-in field – see Fig. 2a-SI. In our calculations, we replace this changing potential inside the barrier with its average value $\bar{U}_b$. So, we use the potential

$$U_{appr}(z)=\begin{cases}0, & z<0\\ \bar{U}_b, & 0<z<b\\ -|eV|, & z>b\end{cases} \qquad \text{(SI-8)}$$

where b is the barrier thickness. The barrier height $\bar{U}_b$ can be written as $\bar{U}_b=\varepsilon_F+W-|eV|/2$ where ε_F is Fermi energy in the metals, W is the work function from the metal into the conduction band of the dielectric (semiconductor) in the barrier.

(2) It is not overly hard to calculate the exact wavefunctions $\psi_L(z)$ and $\psi_R(z)$ for the potential of Eq. (SI-8). Nevertheless, for our purposes it is sufficient to employ the approximated functions ("tunneling functions"), used often in tunneling theory [Payne, M. C., Transfer Hamiltonian description of resonant tunneling, J. Phys. C: Solid State Phys., **1986**, 19**,** 1145-1155]. Namely, $\psi_L(z)$ is calculated for the potential

$$U_L(z)=\begin{cases}0, & z<0\\ \bar{U}_b, & z>0\end{cases} \qquad \text{(SI-9)}$$

but $\psi_R(z)$ is calculated for the potential

$$U_R(z)=\begin{cases}\bar{U}_b, & z<b\\ -|eV|, & z>b\end{cases} \tag{SI-10}$$

Correspondingly, we have the following wave functions:

$$\psi_L(z)=\frac{1}{\sqrt{\mathrm{v}_z}}\begin{cases}e^{+ik_{zL}z}+A_L e^{-ik_{zL}z}, & z<0\\ B_L e^{-\kappa_L z}, & z>0\end{cases} \tag{SI-11}$$

where $\mathrm{v}_z=\hbar k_{zL}/m$, $\dfrac{\hbar^2\kappa_L^2}{2m}=\bar{U}_b-\dfrac{\hbar^2k_{zL}^2}{2m}$, $A_L=\dfrac{k_{zL}-i\kappa_L}{k_{zL}+i\kappa_L}$ and $B_L=\dfrac{2k_{zL}}{k_{zL}+i\kappa_L}$; and

$$\psi_R(z)=\begin{cases}B_R e^{+\kappa_R(z-b)}, & z<b\\ e^{-ik_{zR}(z-b)}+A_R e^{+ik_{zR}(z-b)}, & z>b\end{cases} \tag{SI-12}$$

where

$$\frac{\hbar^2k_{zR}^2}{2m}=\frac{\hbar^2k_{zL}^2}{2m}+|eV|-\hbar\omega,\ \frac{\hbar^2\kappa_R^2}{2m}=\frac{\hbar^2\kappa_L^2}{2m}+\hbar\omega,\ A_R=\frac{k_{zR}-i\kappa_R}{k_{zR}+i\kappa_R}\text{ and }B_R=\frac{2k_{zR}}{k_{zR}+i\kappa_R} \tag{SI-13}$$

The above functions $\psi_L(z)$ and $\psi_R(z)$ must be properly normalized. Namely, the initial function $\psi_L(z)$ is normalized onto the unit flux of probability for electron incident on the barrier, but the wave, describing the incidence of the electron on the barrier in the final function $\psi_R(z)$, is normalized onto unit density of probability [Landau, L. D.; Lifshitz, E. M. *Quantum Mechanics*: *Non Relativistic Theory, Third Edition*; Elsevier Science Ltd., 1977]. Fig. 2a-SI in the paper illustrates the behaviour of the functions.

SI-3. Wave functions $\psi_L(z)$ and $\psi_R(z)$ for double-barrier structure with resonant tunneling

Figure 2b-SI shows the energy diagram of structure with two barriers (dielectric or semiconductor) and quantum well (thin layer of metal or dielectric with forbidden gap, narrower than forbidden gap in surrounding barriers) between them. The Quantum Well (QW) has a resonant level (dark blue), into which the electron can make an SPP-assisted transition (red arrow). This feature results in a substantial increase of the inelastic tunneling with emission of the SPP compared to a single-barrier structure. One should stress that elastic tunneling (green arrow) is nonresonant (as in single barrier structure in Fig. 2a-SI).

In the calculation of the initial electron wave function ψ_L impinges with the velocity v_z the onto the barrier structure from the left (L) metal, we make the same approximations as for the single barrier case:

(A) "tunneling approximation", i.e. we calculate ψ_L for the potential

$$U_L(z)=\begin{cases} 0, & z<-b_1 \\ \bar{U}_{b1} & -b_1<z<0 \\ \bar{U}_{qw} & 0<z<l_{qw} \\ \bar{U}_{b2} & z>l_{qw} \end{cases} \tag{SI-14}$$

where $\bar{U}_{b1}$ ($\bar{U}_{b2}$) is the average potential in barrier 1 (2), and $\bar{U}_{qw}$ is the average potential in QW;

(B) we consider the case of *nonresonant* tunneling, i.e. electron before emitting the SPP is off resonances with levels in the QW, so that we can write for ψ_L:

$$\psi_L = \frac{1}{\sqrt{v_z}} \begin{cases} e^{+ik_{zL}(z+b_1)}, & z < -b_1 \\ e^{-\kappa_{L1}(z+b_1)}, & -b_1 < z < 0 \\ e^{-\kappa_{L1}b_1} \cdot e^{+ik_{Lqw}z}, & 0 < z < l_{qw} \\ e^{-\kappa_{L1}b_1} \cdot e^{+ik_{Lqw}a_2} \cdot e^{-\kappa_{L2}(z-l_{qw})} & z > l_{qw} \end{cases} \tag{SI-15}$$

where $v_z = \hbar k_{zL}/m$, $\frac{\hbar^2\kappa_{L1}^2}{2m} = \bar{U}_{b1} - \frac{\hbar^2 k_{zL}^2}{2m}$, $\frac{\hbar^2\kappa_{L2}^2}{2m} = \bar{U}_{b2} - \frac{\hbar^2 k_{zL}^2}{2m}$, $\frac{\hbar^2 k_{Lqw}^2}{2m} = \frac{\hbar^2 k_{zL}^2}{2m} - \bar{U}_{qw}$. Note that ψ_L is normalized on unit flux of probability in the left metal.

The function ψ_R of an electron after SPP emission is calculated for the potential

$$U_R(z) = \begin{cases} \bar{U}_{b1}, & z < 0 \\ \bar{U}_{qw}, & 0 < z < l_{qw} \\ \bar{U}_{b2} & l_{qw} < z < l_{qw} + b_2 \\ -|eV| & z > l_{qw} + b_2 \end{cases} \tag{SI-16}$$

In the region $z > l_{qw} + b_2$ (right metal), the wave function ψ_R includes the wave $e^{-ik_{zR}z}$ (normalized on unit density of probability) where $k_{zR} = \sqrt{k_{zL}^2 + \frac{2m|eV|}{\hbar^2} - \frac{2m\hbar\omega_{sp}}{\hbar^2}}$. In the QW region ($0 < z < l_{qw}$), the function ψ_R can be obtained (as in optics in consideration of light passing through Fabry-Perot) by summing waves in this region, which arise due to the tunneling of the incident wave through the barrier 2 with the coefficient $t_{\leftarrow}^{(2)}$, and subsequent multiple reflections from the barriers 1 and 2 with the coefficients $r_{\rightarrow}^{(1)}$ and $r_{\leftarrow}^{(2)}$, correspondingly:

$$\psi_R(z)=\frac{t_{\leftarrow}^{(2)}e^{+ik_{Rqw}l_{qw}}}{1-e^{2ik_{Rqw}l_{qw}}r_{\rightarrow}^{(1)}r_{\leftarrow}^{(2)}}\cdot\left(e^{-ik_{Rqw}z}+e^{+ik_{Rqw}z}r_{\rightarrow}^{(1)}\right)\equiv A_{\mathrm{qw}}\cdot\left(e^{-ik_{Rqw}z}+e^{+ik_{Rqw}z}r_{\rightarrow}^{(1)}\right) \qquad \text{(SI-17)}$$

where $\frac{\hbar^2 k_{Rqw}^2}{2m}=\frac{\hbar^2 k_{zL}^2}{2m}-\bar{U}_{qw}-\hbar\omega_{sp}$. For $z<0$, the wave function $\psi_R(z)=A_{qw}\cdot\left(1+r_{\rightarrow}^{(1)}\right)\cdot e^{+\kappa_{R1}z}$.

The above formulas for ψ_R are general, and valid both for an electron at resonance and off resonance with a level in the QW. If the electron energy $E_R=E_L-\hbar\omega_{sp}$ is close to the QW's level E_{QW}, ψ_R can be written approximately as

$$\psi_R(z)=A_{qw}^{res}\cdot\begin{cases}\left(1+r_{\rightarrow}^{(1)}\right)\cdot e^{+\kappa_{R1}z}, & z<0\\ \left(e^{-ik_{Rqw}z}+e^{+ik_{Rqw}z}r_{\rightarrow}^{(1)}\right), & 0<z<l_{qw}\\ \left(e^{-ik_{Rqw}l_{qw}}+e^{+ik_{Rqw}l_{qw}}r_{\rightarrow}^{(1)}\right)\cdot e^{-\kappa_{R2}(z-l_{qw})}, & z>l_{qw}\end{cases} \qquad \text{(SI-18)}$$

where $A_{qw}^{res}=2e^{+\kappa_{R2}b_2-i\delta_R}\cdot\left[1+\left(\frac{E_R-E_{QW}}{\Delta E_{res}}\right)^2\right]^{-1/2}$; $\Delta E_{res}=e^{-2\kappa_{R2}b_2}\cdot\left(E_R-\bar{U}_{qw}\right)/2\pi$ is the width of the level in the QW. Note that here and below we assume that electron is in resonance with fundamental energy level of the QW.

SI-4. Elastic and inelastic current density in structures with resonant tunneling

Using formulas

$$p_{DB,res}^{inelast}\left(\mathrm{v}_z\right)\sim e^{-2\kappa_{L1}b_1}\cdot\frac{E_{QW}-\bar{U}_{qw}}{2\pi\cdot\Delta E_{QW}}\frac{1}{1+\left(\dfrac{E_R-E_{QW}}{\Delta E_{QW}}\right)^2}\cdot\frac{\alpha_{fs}}{4\pi^2\left(1+\varepsilon_d\right)}\cdot\frac{\mathrm{v}_z^2}{c^2}\cdot\frac{\lambda_r^3}{V_{nano}} \tag{SI-19}$$

and

$$p_{DB}^{elast}\sim e^{-2\kappa_{L1}b_1}\cdot e^{-2\kappa_{L2}b_2} \tag{SI-20}$$

in

$$J_z=e\cdot\frac{mk_BT_e}{2\pi^2\hbar^3}\cdot\int_0^{+\infty}dE_z\cdot p\left(E_z\right)\cdot\ln\left(1+\mathrm{e}^{-\frac{E_z-\varepsilon_F}{k_BT_e}}\right), \tag{SI-21}$$

where $E_z=m\mathrm{v}_z^2/2$ is the energy of the incident electron, we get for the current densities due to inelastic tunneling

$$J_{DB,res}^{inel}=e\cdot\frac{m\varepsilon_F}{2\pi^2\hbar^3}\cdot\exp\left[-2\kappa_1\left(E_z^{res}\right)\cdot b_1\right]\cdot\frac{E_{QW}-\bar{U}_{qw}}{2}\cdot\frac{E_z^{res}}{\varepsilon_F}\cdot\eta^{nonres}\cdot\frac{\varepsilon_F-E_z^{res}}{\varepsilon_F} \tag{SI-22}$$

and elastic tunneling

$$J_{DB}^{el}=e\cdot\frac{m\varepsilon_F}{2\pi^2\hbar^3}\cdot\exp\left[-2\cdot\kappa_1^{\varepsilon_F}\cdot b_1-2\cdot\kappa_2^{\varepsilon_F}\cdot b_2\right]\cdot\varepsilon_F\cdot\left(\frac{\&\%}{\varepsilon_F}\right)^2 \tag{SI-23}$$

where $E_z^{res}=\hbar\omega_{sp}+E_{QW}$, $\kappa_1\left(E_z^{res}\right)=\sqrt{2m\left(\bar{U}_{b1}-E_z^{res}\right)/\hbar^2}$, $\kappa_j^{\varepsilon_F}=\sqrt{2m\left(\bar{U}_{bj}-\varepsilon_F\right)/\hbar^2}$ ($j=1,2$),

$\&\%=\left(\dfrac{\kappa_1^{\varepsilon_F}b_1}{\bar{U}_{b1}-\varepsilon_F}+\dfrac{\kappa_2^{\varepsilon_F}b_2}{\bar{U}_{b2}-\varepsilon_F}\right)^{-1}$. In derivation of Eq. (22), we have used the substitution

$\left(\pi\Delta E_{QW}\right)^{-1}\cdot 1\Big/\left\{+\left[\left(E_z-\left(\hbar\omega_{sp}+E_{QW}\right)\right)\Big/\Delta E_{QW}\right]^2\right\}\to\delta\left[E_z-\left(\hbar\omega_{sp}+E_{QW}\right)\right]$. Assuming, that in (SI-23), $E_z^{res}\approx\varepsilon_F$ and $\left(\varepsilon_F-E_z^{res}\right)\Big/\varepsilon_F\sim 1$`, we can estimate the quantum efficiency as

$$\eta_{res}=\frac{\eta_{nonres}}{D_{L2}\left(\varepsilon_F\right)\cdot\frac{2\varepsilon_F}{E_{QW}-\bar{U}_{qw}}\left(\frac{\tilde{\varepsilon}}{\varepsilon_F}\right)^2+\eta_{nonres}} \qquad \text{(SI-24)}$$