# SUPPORTING INFORMATION
# Role of Depolarization Factors in the Evolution of a Dipolar Plasmonic Spectral Line in the Far- and Near-Field Regimes

Mochamad Januar,[†] Bei Liu,[†] Jui-Ching Cheng,[‡] Koji Hatanaka,[¶,§,‖] Hiroaki Misawa,[⊥,#] Hui-Hsin Hsiao,[*,@] and Kou-Chen Liu[*,†,△,∇]

†*Department of Electronic Engineering, Chang Gung University, Taoyuan 33302, Taiwan*

‡*Department of Electronic Engineering, National Taipei University of Technology, Taipei 10608, Taiwan*

¶*Research Center for Applied Sciences, Academia Sinica, Taipei 11529, Taiwan*

§*College of Engineering, Chang Gung University, Taoyuan 33302, Taiwan*

‖*Department of Materials Science and Engineering, National Dong Hwa University, Hualien 97401, Taiwan*

⊥*Research Institute for Electronic Science, Hokkaido University, Sapporo 001-0021, Japan*

#*Center for Emergent Functional Matter Science, National Chiao Tung University, Hsinchu 30010, Taiwan*

@*Institute of Electro-Optical Engineering, National Taiwan Normal University, Taipei 11677, Taiwan*

△*Division of Pediatric Infectious Disease, Department of Pediatrics, Chang Gung Memorial Hospital, Linkou 33305, Taiwan*

∇*Department of Materials Engineering, Ming Chi University of Technology, New Taipei City 24301, Taiwan*

E-mail: hhhsiao@ntnu.edu.tw; jacobliu@mail.cgu.edu.tw

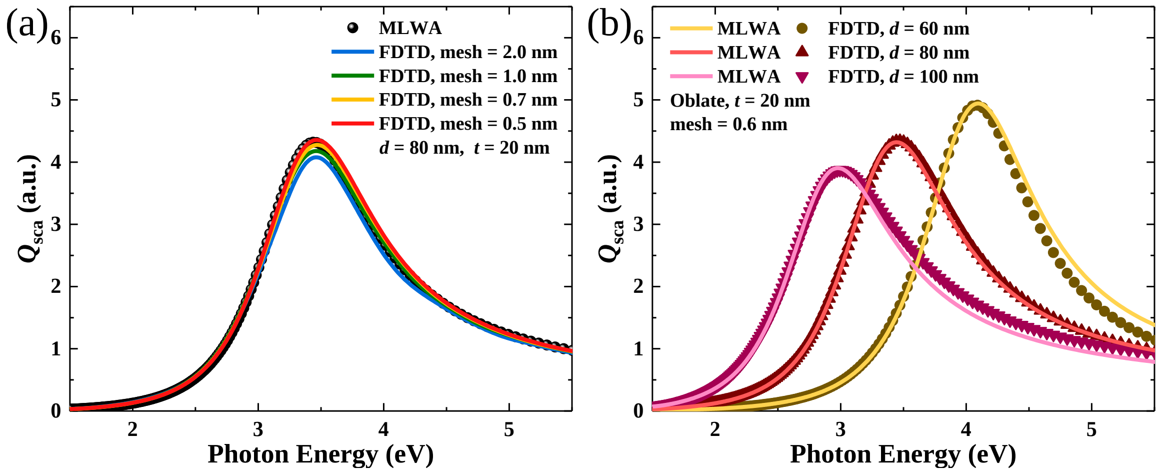


Figure S1: The far-field spectra obtained with the MLWA model (curves) and the numerical full-wave electrodynamics method (symbols) for oblate-shaped particles with different (a) mesh sizes and (b) particle diameters.

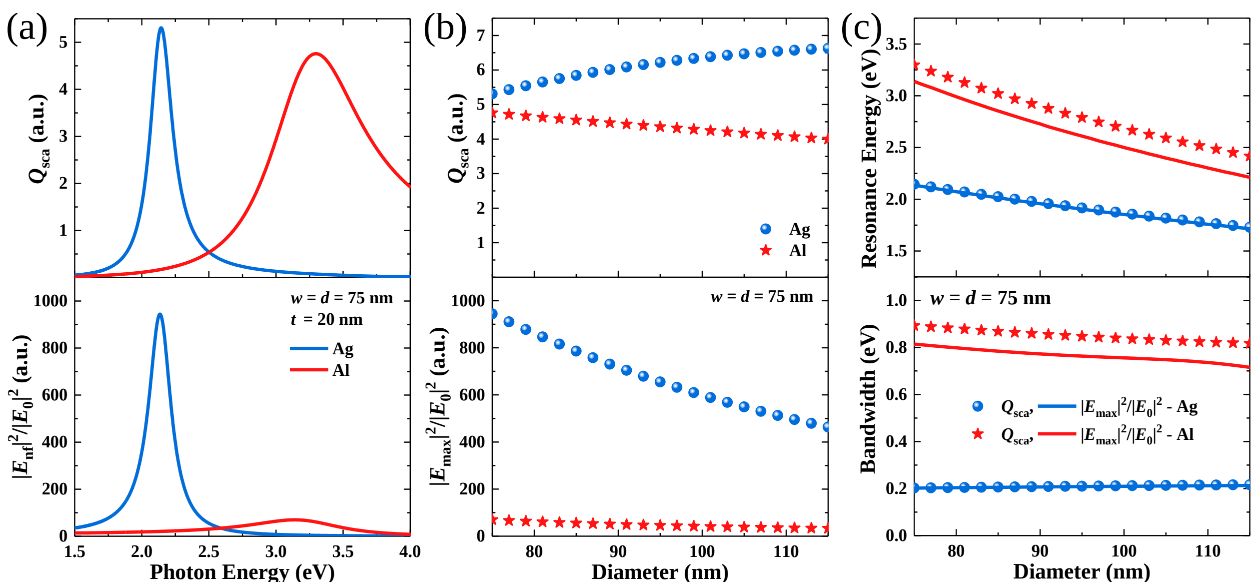


Figure S2: (a) The far-field (upper) and near-field (lower) spectra for Ag and Al oblate ellipsoidal particle. (b) The variation of far-field (upper) and near-field (lower) intensity as a function of diameter, and (c) the corresponding trend of resonant energy (upper) and bandwidth (lower).

In this supplementary material, we elaborate on the derivation of the analytical forms provided in the main paper in greater detail (the expressions in Table 1, *i.e.*, eqs 14 and 15, and eqs 19 to 24). We begin from the expressions for the dipolar polarizability in the MLWA model (refers to eq 8 of the manuscript), which is given by

$$\alpha_{\text{eff}} = V \frac{\epsilon - \epsilon_{\text{m}}}{\epsilon_{\text{m}} + L_{\text{eff}}(\epsilon - \epsilon_{\text{m}})}, \tag{S1}$$

where $L_{\text{eff}}$ is the effective depolarization-factor with the inclusion of the finite wavelength corrections given by

$$L_{\text{eff}}(\omega) = L - L_{\text{dyn}}\omega^2 - iL_{\text{rad}}\omega^3. \tag{S2}$$

Moreover, the Drude model of the metallic dispersion relation is given by[1,2]

$$\epsilon(\omega) = 1 - \frac{\omega_{\text{p}}^2}{\omega^2 + i\gamma\omega}. \tag{S3}$$

To have a simpler form, we modify eq S1 to become

$$\alpha_{\text{eff}} = \frac{V}{\frac{\epsilon_{\text{m}}}{\epsilon - \epsilon_{\text{m}}} + L_{\text{eff}}}. \tag{S4}$$

Then, by defining $\epsilon$ as the Drude dielectric-function in eq S3, one obtains

$$\begin{aligned} \alpha_{\text{eff}} &= \frac{V}{\frac{\epsilon_{\text{m}}}{1 - \frac{\omega_{\text{p}}^2}{\omega^2 + i\gamma\omega} - \epsilon_{\text{m}}} + L_{\text{eff}}} \\ &= \frac{V}{\frac{1}{\frac{1}{\epsilon_{\text{m}}} - 1 - \frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}(\omega^2 + i\gamma\omega)}} + L_{\text{eff}}}. \end{aligned} \tag{S5}$$

For $\omega \ll \omega_{\text{p}}$, we can approach that $\frac{1}{\epsilon_{\text{m}}} - 1 \ll \frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}(\omega^2 + i\gamma\omega)}$. This approach is reasonable because the general interest of the plasmonic effects is between the infrared and the near-ultraviolet regimes, which are far below the plasma frequency of most plasmonic metal materials. Therefore, eq S5 can be simplified as

$$\begin{aligned}
\alpha_{\text{eff}} &= \frac{V}{-\frac{\epsilon_{\text{m}}(\omega^2+i\gamma\omega)}{\omega_{\text{p}}^2}+L_{\text{eff}}} \\
&= V\frac{\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}{\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}L_{\text{eff}}-\omega^2-i\gamma\omega}.
\end{aligned} \tag{S6}$$

Furthermore, inserting $L_{\text{eff}}$ in eq S2 into eq S6, one obtains

$$\alpha_{\text{eff}} = V\frac{\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}{\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}\left(L-L_{\text{dyn}}\omega^2-iL_{\text{rad}}\omega^3\right)-\omega^2-i\gamma\omega}. \tag{S7}$$

Subsequently, the equation can be simplified by regrouping the terms according to the real and imaginary components as

$$\alpha_{\text{eff}} = V\frac{\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}{L\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}-\left(1+L_{\text{dyn}}\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}\right)\omega^2-i\left(\gamma+L_{\text{rad}}\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}\omega^2\right)\omega}. \tag{S8}$$

To clearly see the resonant criterion from the equation, we can multiply both the numerator and denominator of eq S8 by $\frac{1}{1+L_{\text{dyn}}\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}$, then we have

$$\begin{aligned}
\alpha_{\text{eff}} &= V\frac{\frac{\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}{1+L_{\text{dyn}}\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}}{\frac{L\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}{1+L_{\text{dyn}}\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}-\omega^2-i\left(\frac{\gamma+L_{\text{rad}}\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}\omega^2}{1+L_{\text{dyn}}\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}}}\right)\omega} \\
&= V\frac{\frac{\omega_{\text{p}}^2}{\epsilon_{\text{m}}+L_{\text{dyn}}\omega_{\text{p}}^2}}{\frac{L\omega_{\text{p}}^2}{\epsilon_{\text{m}}+L_{\text{dyn}}\omega_{\text{p}}^2}-\omega^2-i\left(\frac{\epsilon_{\text{m}}\gamma+L_{\text{rad}}\omega_{\text{p}}^2\omega^2}{\epsilon_{\text{m}}+L_{\text{dyn}}\omega_{\text{p}}^2}\right)\omega}.
\end{aligned} \tag{S9}$$

At this point, one can see that the pole of the real part of eq S9 defines the resonant frequency of the LSPR response as

$$\omega_{\text{lspr}} \equiv \sqrt{\frac{L\omega_{\text{p}}^2}{\epsilon_{\text{m}}+L_{\text{dyn}}\omega_{\text{p}}^2}}. \tag{S10}$$

Correspondingly, at the resonance (when $\omega = \omega_{\text{lspr}}$), the imaginary term would determine the bandwidth of the spectrum, which can be approached as

$$\Gamma_{\text{lspr}} \equiv \frac{\epsilon_{\text{m}}\gamma + L_{\text{rad}}\omega_{\text{p}}^2\omega_{\text{lspr}}^2}{\epsilon_{\text{m}} + L_{\text{dyn}}\omega_{\text{p}}^2}. \tag{S11}$$

Through these approximations, we can rewrite the expression in eq S9 as

$$\alpha_{\text{eff}} = \frac{V}{L}\frac{\omega_{\text{lspr}}^2}{\omega_{\text{lspr}}^2 - \omega^2 - i\Gamma_{\text{lspr}}\omega}. \tag{S12}$$

Hence, we have arrived at the similar expressions as shown in eqs 14 and 15 in the main text.

The above results can subsequently be used to obtain qualitative descriptions of the magnitudes of the far- and near-field efficiencies through the following expressions (eqs 10 and 11 in the main text)

$$Q_{\text{sca}} = \frac{k^4 \left|\alpha_{\text{eff}}\right|^2}{6\pi^2 \left(\frac{d}{2}\right)^2}, \tag{S13}$$

$$\frac{\left|E_{\text{nf}}\right|^2}{\left|E_0\right|^2} = \left|1 - \frac{L_{\text{eff}}\alpha_{\text{eff}}}{V}\right|^2. \tag{S14}$$

First, we evaluate the expression for the far-field scattering by considering the complex amplitude of the polarizability in eq S12 as

$$\begin{aligned}
\left|\alpha_{\text{eff}}\right|^2 &= \alpha_{\text{eff}}\alpha_{\text{eff}}^* \\
&= \left(\frac{V}{L}\omega_{\text{lspr}}^2\right)^2 \left(\frac{1}{\omega_{\text{lspr}}^2 - \omega^2 - i\Gamma_{\text{lspr}}\omega}\right) \times \left(\frac{1}{\omega_{\text{lspr}}^2 - \omega^2 + i\Gamma_{\text{lspr}}\omega}\right) \\
&= \left(\frac{V}{L}\omega_{\text{lspr}}^2\right)^2 \frac{1}{\left(\omega_{\text{lspr}}^2 - \omega^2\right)^2 + \left(\Gamma_{\text{lspr}}\omega\right)^2}.
\end{aligned} \tag{S15}$$

Here, $\alpha_{\text{eff}}^*$ denotes the complex conjugate of $\alpha_{\text{eff}}$. Furthermore, substituting eq S15 into eq S13 and taking $k = \frac{\omega}{c}$, we get

$$Q_{\text{sca}}\left(\omega\right) = \frac{2}{3\pi^2 c^4}\frac{V^2\omega_{\text{lspr}}^4}{L^2 d^2}\frac{\omega^4}{\left(\omega_{\text{lspr}}^2 - \omega^2\right)^2 + \left(\Gamma_{\text{lspr}}\omega\right)^2}. \tag{S16}$$

This is eq 19 from the main text. As explained in the paper, eq S16 resembles a Lorentzian distribution function centered at $\omega_{\text{lspr}}$ (eq S10) with a full width at half-maximum of $\Gamma_{\text{lspr}}$ (eq S11). By taking $\omega = \omega_{\text{lspr}}$, eq S16 becomes

$$Q_{\text{sca}}\left(\omega_{\text{lspr}}\right) = \frac{2}{3\pi^2 c^4}\frac{V^2}{L^2 d^2}\frac{\omega_{\text{lspr}}^6}{\Gamma_{\text{lspr}}^2}. \tag{S17}$$

This equation gives the proportionality expression for the far-field efficiency at resonance as

$$Q_{\text{sca}}\big|_{\text{res}} \propto \left(\frac{V}{Ld}\frac{\omega_{\text{lspr}}^3}{\Gamma_{\text{lspr}}}\right)^2, \tag{S18}$$

which results in the expression for eq 21 of the main paper.

Second, to deduce the expression for the near-field enhancement, one can substitute eq S12 into eq S14. Since the magnitude of the near-field intensity is usually much larger than unity, as also shown in the main text, eq S14 can be simplified as,

$$\frac{\left|E_{\text{nf}}\right|^2}{\left|E_0\right|^2} \approx \left|\frac{L_{\text{eff}}\alpha_{\text{eff}}}{V}\right|^2. \tag{S19}$$

Hence, we can evaluate the complex-amplitude term as

$$\begin{aligned}
\left|L_{\text{eff}}\alpha_{\text{eff}}\right|^2 &= \left(L_{\text{eff}}\alpha_{\text{eff}}\right)\left(L_{\text{eff}}\alpha_{\text{eff}}\right)^* \\
&= L_{\text{eff}}L_{\text{eff}}^*\alpha_{\text{eff}}\alpha_{\text{eff}}^* \\
&= \left|L_{\text{eff}}\right|^2\left|\alpha_{\text{eff}}\right|^2.
\end{aligned} \tag{S20}$$

Subsequently, inserting this expression into the near-field enhancement in eq S19 and using eq S15 for the $|\alpha_{\text{eff}}|^2$-term, we can obtain

$$\begin{aligned}\frac{|E_{\text{nf}}|^2}{|E_0|^2} &= \frac{1}{V^2}\,|L_{\text{eff}}|^2\,|\alpha_{\text{eff}}|^2 \\ &= \frac{|L_{\text{eff}}|^2}{V^2}\left[\frac{\left(\frac{V}{L}\omega_{\text{lspr}}^2\right)^2}{\left(\omega_{\text{lspr}}^2-\omega^2\right)^2+\left(\Gamma_{\text{lspr}}\omega\right)^2}\right]. \qquad \text{(S21)}\end{aligned}$$

With a little arrangement, this equation gives the qualitative expression for the near-field efficiency shown in eq 20 in the main text as follows

$$\frac{|E_{\text{nf}}|^2}{|E_0|^2}(\omega) = \frac{|L_{\text{eff}}|^2\,\omega_{\text{lspr}}^4}{L^2}\,\frac{1}{\left(\omega_{\text{lspr}}^2-\omega^2\right)^2+\left(\Gamma_{\text{lspr}}\omega\right)^2}. \qquad \text{(S22)}$$

As described in the main text, eq S22 is not a Lorentzian function. The maximum amplitude of the near field is reached at a frequency for which the derivative of the denominator of eq S22 is equal to zero. This approach gives

$$\begin{aligned}\frac{d}{d\omega}\left\{\left(\omega_{\text{lspr}}^2-\omega^2\right)^2+\left(\Gamma_{\text{lspr}}\omega\right)^2\right\} &= 0 \\ -4\omega\left(\omega_{\text{lspr}}^2-\omega^2\right)+2\Gamma_{\text{lspr}}^2\omega &= 0 \\ \left(\omega^2-\omega_{\text{lspr}}^2+\frac{\Gamma_{\text{lspr}}^2}{2}\right)\omega &= 0 \\ \omega^2-\omega_{\text{lspr}}^2+\frac{\Gamma_{\text{lspr}}^2}{2} &= 0. \qquad \text{(S23)}\end{aligned}$$

By taking only the positive component of the square root, this relation gives the effective resonant frequency for the near-field spectra ($\omega_{\text{nf}}$) as

$$\omega_{\text{nf}} = \omega_{\text{lspr}}\sqrt{1-\frac{1}{2}\left(\frac{\Gamma_{\text{lspr}}}{\omega_{\text{lspr}}}\right)^2}. \qquad \text{(S24)}$$

This is eq 22 from the main text. For brevity, we are defining $\Delta_{\text{lspr}} \equiv \sqrt{1-\frac{1}{2}\left(\frac{\Gamma_{\text{lspr}}}{\omega_{\text{lspr}}}\right)^2}$, as shown in Table 1. Therefore, we have

$$\omega_{\text{nf}} = \Delta_{\text{lspr}}\omega_{\text{lspr}}. \tag{S25}$$

Subsequently, by substituting $\omega_{\text{lspr}}$ in this expression back into eq S22, the bandwidth of the near-field spectra can be obtained from

$$\begin{aligned} \frac{|E_{\text{nf}}|^2}{|E_0|^2}(\omega) &= \frac{|L_{\text{eff}}|^2}{L^2}\left(\frac{\omega_{\text{nf}}}{\Delta_{\text{lspr}}}\right)^4 \frac{1}{\left(\left(\frac{\omega_{\text{nf}}}{\Delta_{\text{lspr}}}\right)^2 - \omega^2\right)^2 + (\Gamma_{\text{lspr}}\omega)^2} \\ &= \frac{|L_{\text{eff}}|^2}{L^2}\frac{\omega_{\text{nf}}^4}{\left(\omega_{\text{nf}}^2 - (\Delta_{\text{lspr}}\omega)^2\right)^2 + (\Delta_{\text{lspr}}\Gamma_{\text{lspr}}\Delta_{\text{lspr}}\omega)^2}. \end{aligned} \tag{S26}$$

To simplify the equation, we approached that $\omega' \cong \Delta_{\text{lspr}}\omega$. This way, eq S26 can be modified as

$$\begin{aligned} \frac{|E_{\text{nf}}|^2}{|E_0|^2}(\omega') &= \frac{|L_{\text{eff}}|^2}{L^2}\frac{\omega_{\text{nf}}^4}{(\omega_{\text{nf}}^2 - \omega'^2)^2 + (\Delta_{\text{lspr}}\Gamma_{\text{lspr}}\omega')^2} \\ &= \frac{|L_{\text{eff}}|^2}{L^2}\frac{\omega_{\text{nf}}^4}{(\omega_{\text{nf}}^2 - \omega'^2)^2 + (\Gamma_{\text{nf}}\omega')^2}. \end{aligned} \tag{S27}$$

Hence, the bandwidth of the near-field spectra can be straightforwardly deduced from eq S27 as

$$\begin{aligned} \Gamma_{\text{nf}} &\equiv \Gamma_{\text{lspr}}\Delta_{\text{lspr}} \\ &= \Gamma_{\text{lspr}}\sqrt{1 - \frac{1}{2}\left(\frac{\Gamma_{\text{lspr}}}{\omega_{\text{lspr}}}\right)^2}. \end{aligned} \tag{S28}$$

This is eq 23 from the main text. Finally, by taking $\omega' = \omega_{\text{nf}}$ and returning both eqs S24 and S28 into eq S27, we can obtain the intensity of the local field at resonance as

$$\left.\frac{|E_{\text{nf}}|^2}{|E_0|^2}\right|_{\text{res}} = \left(\frac{|L_{\text{eff}}|}{L}\frac{\omega_{\text{lspr}}}{\Gamma_{\text{lspr}}}\right)^2. \tag{S29}$$

This is eq 24 from the main text.

## References

(1) Biagioni, P.; Huang, J.-S.; Hecht, B. Nanoantennas for Visible and Infrared Radiation. *Reports on Progress in Physics* **2012**, *75*, 024402.

(2) Rakić, A. D. Algorithm for the Determination of Intrinsic Optical Constants of Metal Films: Application to Aluminum. *Appl. Opt.* **1995**, *34*, 4755–4767.