

Supporting Information

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SI Text

Boundary Equations for the Asymmetric Shell Method. Following Eq. 14, we have the following eight boundary equations:

$$a_{lm}^{in} \left(\frac{R_1}{R_0} \right)^l = b_{lm}^- - \frac{R_1 + \Delta d}{R_0} \left\{ \begin{aligned} & [(R_1/R_0 + R_0/R_1)^2 + 4(A_l^{m2} + A_{l+1}^{m2})] b_{lm}^+ \\ & -4(R_1/R_0 + R_0/R_1)(b_{l+1m}^+ A_{l+1}^m + b_{l-1m}^+ A_l^m) \\ & + 4(b_{l+2m}^+ A_{l+2}^m A_{l+1}^m + b_{l-2m}^+ A_{l-1}^m A_l^m) \end{aligned} \right\}, \quad [S1]$$

$$\begin{aligned} & \frac{\varepsilon_m R_1^{l+1}}{C_1 R_0^{l-1}} \left[a_{lm}^{in} \left(l+1 + l \frac{R_0^2}{R_1^2} \right) - a_{l-1m}^{in} (2l-1) \frac{R_0^2}{R_1^2} A_l^m - a_{l+1m}^{in} (2l+3) A_{l+1}^m \right] \\ & = \Delta d \left\{ \begin{aligned} & [(R_1/R_0 + R_0/R_1)^2 + 4(A_l^{m2} + A_{l+1}^{m2})] b_{lm}^+ \\ & -4(R_1/R_0 + R_0/R_1)(b_{l+1m}^+ A_{l+1}^m + b_{l-1m}^+ A_l^m) + 4(b_{l+2m}^+ A_{l+2}^m A_{l+1}^m + b_{l-2m}^+ A_{l-1}^m A_l^m) \end{aligned} \right\}, \quad [S2] \end{aligned}$$

$$(a_{lm}^+ + a_{lm}^{s+}) \left(\frac{R_1}{R_0} \right)^l + a_{lm}^- \left(\frac{R_1}{R_0} \right)^{-(l+1)} = b_{lm}^- - \frac{R_1}{R_0} \left\{ \begin{aligned} & [(R_1/R_0 + R_0/R_1)^2 + 4(A_l^{m2} + A_{l+1}^{m2})] b_{lm}^+ \\ & -4(R_1/R_0 + R_0/R_1)(b_{l+1m}^+ A_{l+1}^m + b_{l-1m}^+ A_l^m) + 4(b_{l+2m}^+ A_{l+2}^m A_{l+1}^m + b_{l-2m}^+ A_{l-1}^m A_l^m) \end{aligned} \right\}, \quad [S3]$$

$$\begin{aligned} & \frac{R_1^{l+1}}{C_1 R_0^{l-1}} \left[\begin{aligned} & (a_{lm}^+ + a_{lm}^{s+}) \left(l+1 + l \frac{R_0^2}{R_1^2} \right) - (a_{l-1,m}^+ + a_{l-1,m}^{s+}) (2l-1) A_l^m \frac{R_0^2}{R_1^2} \\ & - (a_{l+1,m}^+ + a_{l+1,m}^{s+}) (2l+3) A_{l+1}^m - a_{lm}^- \left(l+1 + l \frac{R_1^2}{R_0^2} \right) \frac{R_0^{2l+3}}{R_1^{2l+3}} \\ & + (2l-1) A_l^m a_{l-1,m}^- \frac{R_0^{2l+1}}{R_1^{2l+1}} + a_{l+1,m}^- (2l+3) A_{l+1}^m \frac{R_0^{2l+3}}{R_1^{2l+3}} \end{aligned} \right] \\ & = \Delta d \left\{ \begin{aligned} & [(R_1/R_0 + R_0/R_1)^2 + 4(A_l^{m2} + A_{l+1}^{m2})] b_{lm}^+ \\ & -4(R_1/R_0 + R_0/R_1)(b_{l+1m}^+ A_{l+1}^m + b_{l-1m}^+ A_l^m) + 4(b_{l+2m}^+ A_{l+2}^m A_{l+1}^m + b_{l-2m}^+ A_{l-1}^m A_l^m) \end{aligned} \right\}, \quad [S4] \end{aligned}$$

$$a_{lm}^+ \left(\frac{R_2}{R_0} \right)^l + (a_{lm}^- + a_{lm}^{s-}) \left(\frac{R_2}{R_0} \right)^{-(l+1)} = f_{lm}^- - \frac{R_2}{R_0} \left\{ \begin{aligned} & [(R_2/R_0 + R_0/R_2)^2 + 4(A_l^{m2} + A_{l+1}^{m2})] f_{lm}^+ \\ & -4(R_2/R_0 + R_0/R_2)(f_{l+1m}^+ A_{l+1}^m + f_{l-1m}^+ A_l^m) + 4(f_{l+2m}^+ A_{l+2}^m A_{l+1}^m + f_{l-2m}^+ A_{l-1}^m A_l^m) \end{aligned} \right\}, \quad [S5]$$

$$\begin{aligned} & -\frac{R_0^{l+4}}{C_2 R_2^{l+2}} \left[\begin{aligned} & (a_{lm}^- + a_{lm}^{s-}) \left(l+1 + l \frac{R_2^2}{R_0^2} \right) - (a_{l-1,m}^- + a_{l-1,m}^{s-}) (2l-1) A_l^m \frac{R_2^2}{R_0^2} \\ & - (a_{l+1,m}^- + a_{l+1,m}^{s-}) (2l+3) A_{l+1}^m - a_{lm}^+ \left(l+1 + l \frac{R_0^2}{R_2^2} \right) \frac{R_2^{2l+3}}{R_0^{2l+3}} \\ & + a_{l-1,m}^+ (2l-1) A_l^m \frac{R_2^{2l+1}}{R_0^{2l+1}} + a_{l+1,m}^+ (2l+3) A_{l+1}^m \frac{R_2^{2l+3}}{R_0^{2l+3}} \end{aligned} \right] \\ & = \Delta d \left\{ \begin{aligned} & [(R_2/R_0 + R_0/R_2)^2 + 4(A_l^{m2} + A_{l+1}^{m2})] f_{lm}^+ \\ & -4(R_2/R_0 + R_0/R_2)(f_{l+1m}^+ A_{l+1}^m + f_{l-1m}^+ A_l^m) + 4(f_{l+2m}^+ A_{l+2}^m A_{l+1}^m + f_{l-2m}^+ A_{l-1}^m A_l^m) \end{aligned} \right\}, \quad [S6] \end{aligned}$$

$$a_{lm}^{out} \left(\frac{R_2}{R_0} \right)^{-(l+1)} = f_{lm}^- - \frac{R_2 - \Delta d}{R_0} \left\{ \begin{aligned} & [(R_2/R_0 + R_0/R_2)^2 + 4(A_l^{m2} + A_{l+1}^{m2})] f_{lm}^+ \\ & -4(R_2/R_0 + R_0/R_2)(f_{l+1m}^+ A_{l+1}^m + f_{l-1m}^+ A_l^m) \\ & + 4(f_{l+2m}^+ A_{l+2}^m A_{l+1}^m + f_{l-2m}^+ A_{l-1}^m A_l^m) \end{aligned} \right\}, \quad [S7]$$

$$\begin{aligned}
& -\varepsilon_m \frac{R_0^{l+4}}{C_2 R_2^{l+2}} \left[a_{lm}^{out} \left(l+1 + l \frac{R_2}{R_0} \right) - a_{l-1m}^{out} (2l-1) A_l^m \frac{R_2}{R_0} - a_{l+1m}^{out} (2l+3) A_{l+1}^m \right] \\
& = \Delta d \left\{ \left[(R_2/R_0 + R_0/R_2)^2 + 4(A_l^{m2} + A_{l+1}^{m2}) \right] f_{lm}^+ \right. \\
& \quad \left. - 4(R_2/R_0 + R_0/R_2) (f_{l+1m}^+ A_{l+1}^m + f_{l-1m}^+ A_l^m) + 4(f_{l+2m}^+ A_{l+2}^m A_{l+1}^m + f_{l-2m}^+ A_{l-1}^m A_l^m) \right\}, \quad [\text{S8}]
\end{aligned}$$

Eliminating b_{lm}^- and f_{lm}^- in Eqs. S1, S3, S5, and S7 yields

$$\begin{aligned}
(a_{lm}^+ + a_{lm}^{s+}) \left(\frac{R_1}{R_0} \right)^l + a_{lm}^- \left(\frac{R_1}{R_0} \right)^{-(l+1)} - a_{lm}^{in} \left(\frac{R_1}{R_0} \right)^l &= \frac{4\Delta d}{R_0} (b_{l+2m}^+ A_{l+2}^m A_{l+1}^m + b_{l-2m}^+ A_{l-1}^m A_l^m) - \frac{4\Delta d}{R_0} \left(\frac{R_0}{R_1} + \frac{R_1}{R_0} \right) (b_{l+1m}^+ A_{l+1}^m + b_{l-1m}^+ A_l^m) \\
& \quad + \frac{\Delta d}{R_0} \left[\left(\frac{R_1}{R_0} + \frac{R_0}{R_1} \right)^2 + 4(A_l^{m2} + A_{l+1}^{m2}) \right] b_{lm}^+, \quad [\text{S9}]
\end{aligned}$$

$$\begin{aligned}
(a_{lm}^- + a_{lm}^{s-}) \left(\frac{R_2}{R_0} \right)^{-(l+1)} + a_{lm}^+ \left(\frac{R_2}{R_0} \right)^l - a_{lm}^{out} \left(\frac{R_2}{R_0} \right)^{-(l+1)} &= -\frac{4\Delta d}{R_0} (f_{l+2m}^+ A_{l+2}^m A_{l+1}^m + f_{l-2m}^+ A_{l-1}^m A_l^m) + \frac{4\Delta d}{R_0} \left(\frac{R_0}{R_2} + \frac{R_2}{R_0} \right) (f_{l+1m}^+ A_{l+1}^m + f_{l-1m}^+ A_l^m) \\
& \quad - \frac{\Delta d}{R_0} \left[\left(\frac{R_2}{R_0} + \frac{R_0}{R_2} \right)^2 + 4(A_l^{m2} + A_{l+1}^{m2}) \right] f_{lm}^+. \quad [\text{S10}]
\end{aligned}$$

Rewrite Eqs. S2, S4, S6, S8, S9, and S10 in tensor's form

$$\frac{\varepsilon_m R_0^2}{C_1} \mathbf{V}^{++} \mathbf{T}^{++} \mathbf{a}^{in} = \mathbf{U}^{++} \mathbf{U}^{++} \mathbf{b}^+ \Delta d, \quad [\text{S11}]$$

$$\frac{R_0^2}{C_1} [\mathbf{V}^{++} \mathbf{T}^{++} (\mathbf{a}^{S+} + \mathbf{a}^+) - \mathbf{V}^{+-} \mathbf{T}^{+-} \mathbf{a}^-] = \mathbf{U}^{++} \mathbf{U}^{++} \mathbf{b}^+ \Delta d, \quad [\text{S12}]$$

$$-\frac{\varepsilon_m R_0^3}{C_2 R_2} \mathbf{V}^{--} \mathbf{T}^{--} \mathbf{a}^{out} = \mathbf{U}^{--} \mathbf{U}^{--} \mathbf{f}^+ \Delta d, \quad [\text{S13}]$$

$$-\frac{R_0^3}{C_2 R_2} [\mathbf{V}^{--} \mathbf{T}^{--} (\mathbf{a}^{S-} + \mathbf{a}^-) - \mathbf{V}^{-+} \mathbf{T}^{-+} \mathbf{a}^+] = \mathbf{U}^{--} \mathbf{U}^{--} \mathbf{f}^+ \Delta d, \quad [\text{S14}]$$

$$\mathbf{T}^{++} (\mathbf{a}^{S+} + \mathbf{a}^+ - \mathbf{a}^{in}) + \mathbf{T}^{+-} \mathbf{a}^- = \frac{R_1}{R_0^2} \mathbf{U}^{++} \mathbf{U}^{++} \mathbf{b}^+ \Delta d, \quad [\text{S15}]$$

$$\mathbf{T}^{--} (\mathbf{a}^{S-} + \mathbf{a}^- - \mathbf{a}^{out}) + \mathbf{T}^{-+} \mathbf{a}^+ = -\frac{1}{R_0} \mathbf{U}^{--} \mathbf{U}^{--} \mathbf{f}^+ \Delta d, \quad [\text{S16}]$$

where \mathbf{T}^{++} , \mathbf{T}^{+-} , \mathbf{T}^{--} , and \mathbf{T}^{-+} are diagonal matrices defined as

$$\tilde{T}_l^{++} = +\delta_{ll'} \left(\frac{R_1}{R_0} \right)^{l+1}, \quad \tilde{T}_l^{+-} = +\delta_{ll'} \left(\frac{R_1}{R_0} \right)^{-l},$$

$$\tilde{T}_l^{--} = +\delta_{ll'} \left(\frac{R_0}{R_2} \right)^{l+1}, \quad \tilde{T}_l^{-+} = +\delta_{ll'} \left(\frac{R_0}{R_2} \right)^{-l},$$

\mathbf{U}^{++} , \mathbf{U}^{--} , \mathbf{V}^{++} , \mathbf{V}^{+-} , \mathbf{V}^{--} , and \mathbf{V}^{-+} are tridiagonal matrices defined as

$$\tilde{U}_l^{++} = +\delta_{ll'} \left(\frac{R_1}{R_0} + \frac{R_0}{R_1} \right) - 2(\delta_{l'+1} A_{l'+1}^m + \delta_{l'-1} A_{l'-1}^m),$$

$$\tilde{U}_l^{--} = +\delta_{ll'} \left(\frac{R_2}{R_0} + \frac{R_0}{R_2} \right) - 2(\delta_{l'+1} A_{l'+1}^m + \delta_{l'-1} A_{l'-1}^m),$$

$$\begin{aligned}\tilde{V}_{l'}^{++} &= +\delta_{l'} \left[l' \left(1 + \frac{R_1^{-2}}{R_0^{-2}} \right) + 1 \right] - (\delta_{l'+1} A_{l'+1}^m + \delta_{l'-1} A_{l'}^m) (2l' + 1) \frac{R_1^{-1}}{R_0^{-1}}, \\ \tilde{V}_{l'}^{+-} &= +\delta_{l'} \left[l' \left(1 + \frac{R_1^{-2}}{R_0^{-2}} \right) + \frac{R_1^{-2}}{R_0^{-2}} \right] - (\delta_{l'+1} A_{l'+1}^m + \delta_{l'-1} A_{l'}^m) (2l' + 1) \frac{R_1^{-1}}{R_0^{-1}}, \\ \tilde{V}_{l'}^{--} &= +\delta_{l'} \left[l' \left(1 + \frac{R_0^{-2}}{R_2^{-2}} \right) + 1 \right] - (\delta_{l'+1} A_{l'+1}^m + \delta_{l'-1} A_{l'}^m) (2l' + 1) \frac{R_0^{-1}}{R_2^{-1}}, \\ \tilde{V}_{l'}^{-+} &= +\delta_{l'} \left[l' \left(1 + \frac{R_0^{-2}}{R_2^{-2}} \right) + \frac{R_0^{-2}}{R_2^{-2}} \right] - (\delta_{l'+1} A_{l'+1}^m + \delta_{l'-1} A_{l'}^m) (2l' + 1) \frac{R_0^{-1}}{R_2^{-1}}.\end{aligned}$$

Eliminating $\mathbf{U}^{++}\mathbf{U}^{++}\mathbf{b}^+\Delta d$ and $\mathbf{U}^{--}\mathbf{U}^{--}\mathbf{f}^+\Delta d$ in Eqs. S11–S16 yields

$$\varepsilon_m \mathbf{V}^{++} \mathbf{T}^{++} \mathbf{a}^{in} = \mathbf{V}^{++} \mathbf{T}^{++} (\mathbf{a}^{S+} + \mathbf{a}^+) - \mathbf{V}^{+-} \mathbf{T}^{+-} \mathbf{a}^-, \quad [\text{S17}]$$

$$\left(\frac{\varepsilon_m R_1}{C_1} \mathbf{V}^{++} + \mathbf{I} \right) \mathbf{T}^{++} \mathbf{a}^{in} = \mathbf{T}^{++} (\mathbf{a}^{S+} + \mathbf{a}^+) + \mathbf{T}^{+-} \mathbf{a}^-, \quad [\text{S18}]$$

$$\varepsilon_m \mathbf{V}^{--} \mathbf{T}^{--} \mathbf{a}^{out} = \mathbf{V}^{--} \mathbf{T}^{--} (\mathbf{a}^{S-} + \mathbf{a}^-) - \mathbf{V}^{-+} \mathbf{T}^{-+} \mathbf{a}^+, \quad [\text{S19}]$$

$$\left(\frac{\varepsilon_m R_0^2}{C_2 R_2} \mathbf{V}^{--} + \mathbf{I} \right) \mathbf{T}^{--} \mathbf{a}^{out} = \mathbf{T}^{--} (\mathbf{a}^{S-} + \mathbf{a}^-) + \mathbf{T}^{-+} \mathbf{a}^+. \quad [\text{S20}]$$

Eliminating $\mathbf{T}^{++}\mathbf{a}^{in}$ and $\mathbf{T}^{--}\mathbf{a}^{out}$ gives

$$\mathbf{V}^{++} \left[(\varepsilon_m - 1) \mathbf{I} - \frac{\varepsilon_m R_1}{C_1} \mathbf{V}^{++} \right] \mathbf{T}^{++} (\mathbf{a}^{S+} + \mathbf{a}^+) + \left[\left(\frac{\varepsilon_m R_1}{C_1} \mathbf{V}^{++} + \mathbf{I} \right) \mathbf{V}^{+-} + \varepsilon_m \mathbf{V}^{++} \right] \mathbf{T}^{+-} \mathbf{a}^- = 0, \quad [\text{S21}]$$

$$\mathbf{V}^{--} \left[(\varepsilon_m - 1) \mathbf{I} - \frac{\varepsilon_m R_0^2}{C_2 R_2} \mathbf{V}^{--} \right] \mathbf{T}^{--} (\mathbf{a}^{S-} + \mathbf{a}^-) + \left[\left(\frac{\varepsilon_m R_0^2}{C_2 R_2} \mathbf{V}^{--} + \mathbf{I} \right) \mathbf{V}^{-+} + \varepsilon_m \mathbf{V}^{--} \right] \mathbf{T}^{-+} \mathbf{a}^+ = 0. \quad [\text{S22}]$$

Note that \mathbf{V}^{+-} (\mathbf{V}^{-+}) and \mathbf{V}^{++} (\mathbf{V}^{--}) have the following relations:

$$\mathbf{V}^{+-} = \mathbf{V}^{++} + \left(\frac{R_0^2}{R_1^2} - 1 \right) \mathbf{I}, \quad [\text{S23}]$$

$$\mathbf{V}^{-+} = \mathbf{V}^{--} + \left(\frac{R_2^2}{R_0^2} - 1 \right) \mathbf{I}. \quad [\text{S24}]$$

Substituting Eqs. S23 and S24 into Eqs. S21 and S22 yields

$$\mathbf{V}^{++} \left[\mathbf{I} - \frac{\varepsilon_m R_1}{(\varepsilon_m - 1) C_1} \mathbf{V}^{++} \right] \mathbf{T}^{++} (\mathbf{a}^{S+} + \mathbf{a}^+) + \left[\mathbf{V}^{++} e^{-\alpha} + \frac{R_0^2 - R_1^2}{(\varepsilon_m - 1) R_1^2} \mathbf{I} + \frac{\varepsilon_m R_1}{(\varepsilon_m - 1) C_1} \left(\mathbf{V}^{++} + \frac{R_0^2 - R_1^2}{R_1^2} \mathbf{I} \right) \mathbf{V}^{++} \right] \mathbf{T}^{+-} \mathbf{a}^- = 0, \quad [\text{S25}]$$

$$\mathbf{V}^{--} \left[\mathbf{I} - \frac{\varepsilon_m R_0^2}{(\varepsilon_m - 1) C_2 R_2} \mathbf{V}^{--} \right] \mathbf{T}^{--} (\mathbf{a}^{S-} + \mathbf{a}^-) + \left[\mathbf{V}^{--} e^{-\alpha} + \frac{R_2^2 - R_0^2}{(\varepsilon_m - 1) R_0^2} \mathbf{I} + \frac{\varepsilon_m R_0^2}{(\varepsilon_m - 1) C_2 R_2} \left(\mathbf{V}^{--} + \frac{R_2^2 - R_0^2}{R_0^2} \mathbf{I} \right) \mathbf{V}^{--} \right] \mathbf{T}^{-+} \mathbf{a}^+ = 0, \quad [\text{S26}]$$

where we introduced $\alpha = \ln\left(\frac{\varepsilon_m - 1}{\varepsilon_m + 1}\right)$.

Maximum Field Enhancements at the Center of the Gap. The field enhancements (for different geometries and materials) calculated with the asymmetric shell method are given in Figs. S1 and S2.

Fitting Parameters for Gold Permittivity. We simultaneously fit the Olmon et al. (<4.13 eV) (1) and Palik (>4.6 eV) (2) data for gold using the Drude model plus 10 Lorentz terms

$$\varepsilon(\omega) = 1 - \frac{\omega_0^2}{\omega(\omega + i\gamma_0)} - \sum_{j=1}^{10} \frac{f_j \omega_j^2}{\omega^2 - \omega_j^2 + i\gamma_j \omega} \quad [\text{S27}]$$

The fitting parameters are listed in Table S1. The fitting and the Olmon and Palik experimental results are shown in Fig. S3.

Fitting Parameter for Silicon Permittivity. The Palik data for silicon (2) are fitted by using four modified Lorentz terms

$$\varepsilon(\omega) = 1 - \sum_{j=1}^4 \frac{f_j \omega_j^2 - i\gamma_j \omega}{\omega^2 - \omega_j^2 + i\gamma_j \omega} \quad [\text{S28}]$$

The fitting parameters are listed in Table S2. The fitting results and the Palik data are shown in Fig. S4.

1. Olmon RL, et al. (2012) Optical dielectric function of gold. *Phys Rev B* 86(23):235147.
2. Palik EW (1985) *Handbook of Optical Constants of Solids I* (Academic, San Diego).

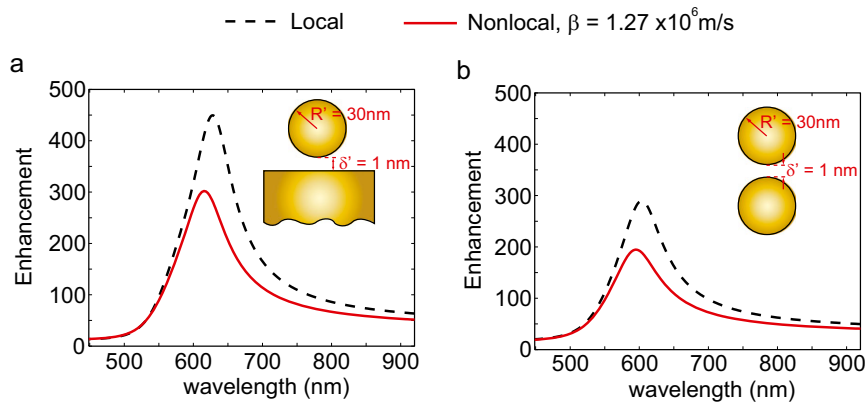


Fig. S1. Maximum field enhancements at the center of the gap for (A) a 30-nm-radius silver on top of a silver plane and (B) two silver spheres of equal radius 30 nm. In both cases, the separation between the two objects is set as 1 nm.

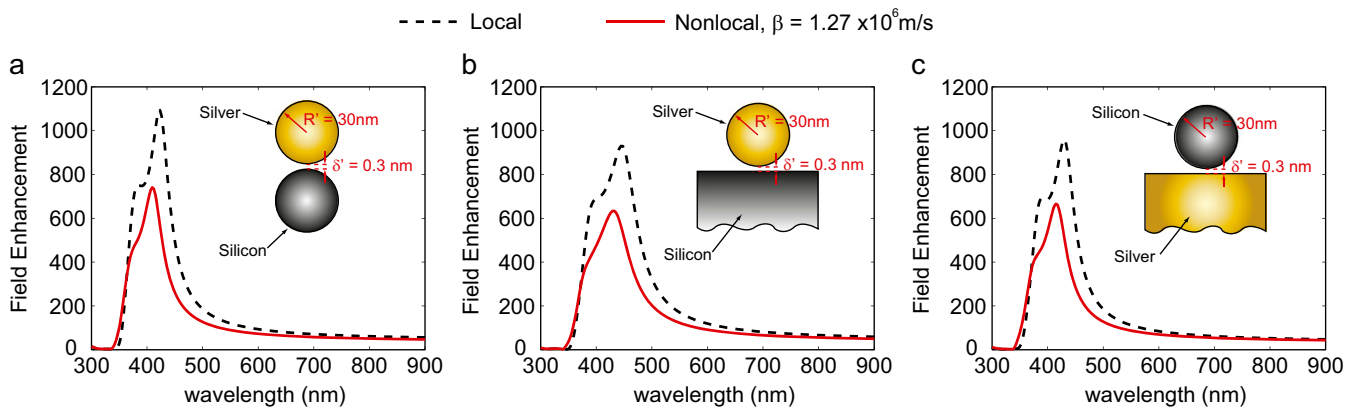


Fig. S2. Maximum field enhancements at the center of the gap for hybrid plasmonic structures (A) between a silver and a silicon spheres of equal radius 30 nm, separated by a 0.3-nm gap; (B) a 30-nm-radius silver sphere placed 0.3 nm above a silicon plane; and (C) a 30-nm-radius silicon sphere placed 0.3 nm above a silver plane.

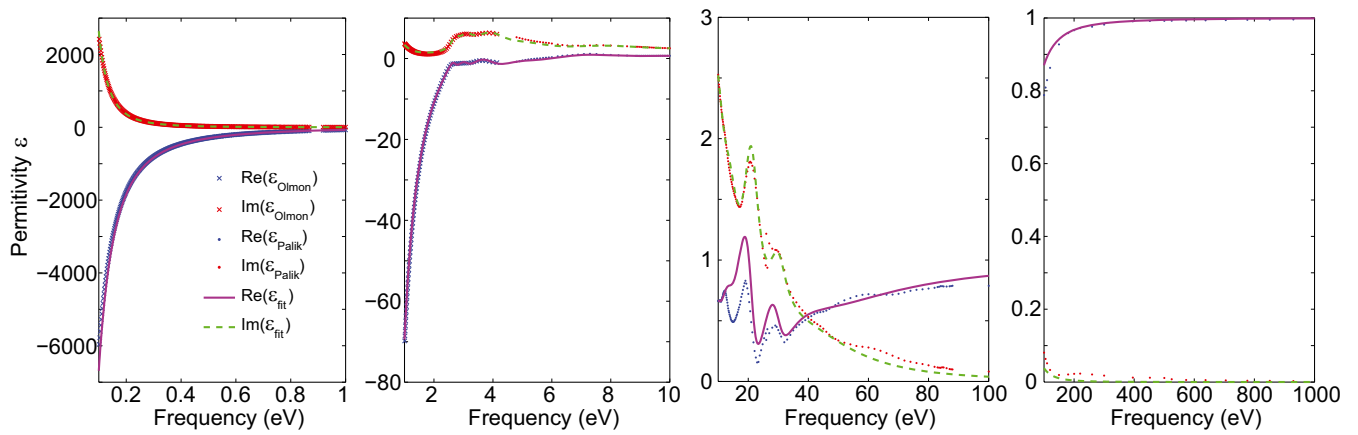


Fig. S3. Fitting results and the Olmon-Palik data for gold. Solid and dashed lines are the real and imaginary parts of the fitted permittivity, respectively. Crosses and dots show the Olmon et al. (1) and Palik experimental data (2), respectively. Different panel corresponds to different frequency ranges.

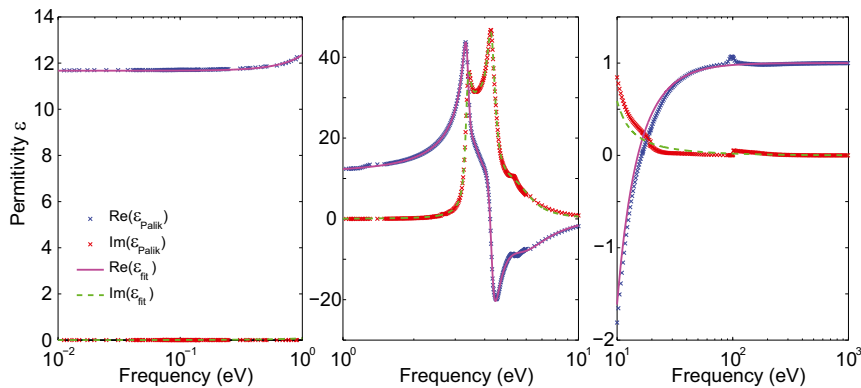


Fig. S4. Fitting results and the Palik data for silicon. Solid and dashed lines are the real and imaginary parts of the fitted permittivity, respectively. Crosses show the Palik data (2). Different panels correspond to different frequency ranges.

Table S1. Fitting parameters for gold

j	f_j	$\omega_j(\text{eV})$	$\gamma_j(\text{eV})$
0	—	8.7663	0.039299
1	0.2975	2.7260	0.3274
2	0.6061	3.1091	0.6066
3	1.4782	3.9415	1.3318
4	0.9284	5.2526	2.3931
5	0.8600	7.9749	3.7913
6	0.4280	10.4650	4.9667
7	0.5323	14.1798	7.8967
8	0.3951	21.2070	6.2030
9	0.1167	30.0898	6.9297
10	0.3728	42.9033	43.5640

Table S2. Fitting parameters for silicon

j	f_j	$\omega_j(\text{eV})$	$\gamma_j(\text{eV})$	$\gamma'_j(\text{eV})$
1	1.3612	3.3779	0.2341	3.3563
2	3.9217	3.6836	0.9397	8.2837
3	2.9488	4.2882	0.4417	-3.7981
4	2.4383	5.5966	2.6091	-5.8775