Supplementary Information

Radiation Engineering of Optical Antennas for Maximum Field Enhancement

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Section I: Derivation of the field enhancement expression for optical antennas using coupled mode theory (CMT)

The mode amplitude of an optical antenna with the effective aperture A_c, illuminated by an excitation beam of spot size A_i, is given from CMT as[1]:

$$\frac{da}{dt} = j\omega_0 a - \frac{1}{2}\left(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{abs}}\right)a + \sqrt{\frac{A_c / A_i}{\tau_{rad}}}\, s_+ \qquad \text{(S1)}$$

where a and $s+$ are the mode amplitude in the cavity and the wave amplitude traveling toward the resonator, respectively, ω_0 is the resonance frequency of the cavity, and $1/\tau_{rad}$ and $1/\tau_{abs}$ express the rate of decay due to radiation and absorption, respectively. With the excitation source of frequency ω, we have:

$$a = \frac{\sqrt{\frac{A_c / A_i}{\tau_{rad}}} s_+}{j(\omega - \omega_0) + \frac{1}{2}(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{abs}})} \tag{S2}$$

Using $|s_+|^2 = \frac{1}{2}\varepsilon_0 E_i^{\,2} c A_i$, the energy stored in the cavity on resonance can be calculated:

$$|a|^2 = \frac{\frac{A_c / A_i}{\tau_{rad}}|s_+|^2}{\frac{1}{4}(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{abs}})^2} = \frac{\frac{A_c / A_i}{\tau_{rad}} \cdot \frac{1}{2}\varepsilon_0 |E_i|^2 c A_i}{\frac{1}{4}(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{abs}})^2} = \frac{1}{2}\varepsilon_0 |E_{loc}|^2 V_{eff} \tag{S3}$$

where E_{loc} and E_i are the local field amplitude at high field region of an optical antenna and the field amplitude of incoming excitation beam, respectively. With the relation $1/\tau = \omega/Q$, we finally get the field enhancement on resonance[2]:

$$\frac{|E_{loc}|^2}{|E_i|^2} = \frac{2A_c\lambda_{res}}{\pi}\frac{Q}{Q_{rad}}\frac{Q}{V_{eff}} \tag{S4}$$

<u>Section II: Derivation of the optimum quality factor condition for optical antennas</u>

We can find the condition for maximum field enhancement by equating derivative of field enhancement equation with respect to Q_{rad} to zero. Since the effective aperture (A_c), effective mode volume (V_{eff}), and the Q_{abs} vary slowly compared to Q_{rad} (see section III in supplementary information and **Fig. 2c**). Then, we have:

$$\begin{aligned}
\frac{\partial}{\partial Q_{rad}}\left(\frac{|E_{loc}|^2}{|E_i|^2}\right) &= \frac{\partial}{\partial Q_{rad}}\left(\frac{2A_c\lambda_{res}}{\pi V_{eff}}\frac{Q^2}{Q_{rad}}\right) \\
&= \frac{2A_c\lambda_{res}}{\pi V_{eff}}\frac{\partial}{\partial Q_{rad}}\frac{1}{\left(Q_{rad}^{-1} + Q_{abs}^{-1}\right)^2 Q_{rad}} \\
&= \frac{2A_c\lambda_{res}}{\pi V_{eff}}\frac{\partial}{\partial Q_{rad}}\frac{Q_{rad}Q_{abs}^2}{\left(Q_{rad} + Q_{abs}\right)^2} \\
&= \frac{2A_c\lambda_{res}}{\pi V_{eff}}\frac{Q_{abs}^2\left(Q_{rad} + Q_{abs}\right)\left(Q_{rad} - Q_{abs}\right)}{\left(Q_{rad} + Q_{abs}\right)^4} \\
&= \frac{2A_c\lambda_{res}}{\pi V_{eff}}\frac{Q_{abs}^2\left(Q_{rad} - Q_{abs}\right)}{\left(Q_{rad} + Q_{abs}\right)^3} = 0
\end{aligned} \tag{S5}$$

Therefore, the maximum field enhancement condition is achieved when Q_{rad} becomes equal to Q_{abs}:

$$Q_{rad} = Q_{abs} \tag{S6}$$

Section III: Effective aperture and effective mode volume of dipole antenna array on ground plane

It has been shown that the complete (normalized) pattern of an array antenna is

$$F(\theta, \phi) = g_a(\theta, \phi) f(\theta, \phi) \tag{S7}$$

where $g_a(\theta, \varphi)$ is the normalized pattern of a single element antenna of the array (the element pattern) and $f(\theta, \varphi)$ is the normalized array factor ("Antenna theory and design" by Stutzman and Thiele). The total pattern is dominated by the array factor if the element pattern is much broader than the array factor and the main beams are aligned. Increasing the array size by adding antenna elements narrows the beamwidth of the array factor and increases the directivity ($D = 4\pi \Big/ \iint |F(\theta, \phi)|^2 d\Omega$). If the antenna array is infinite, the beamwidth of the array factor becomes extremely narrow and the total radiation pattern becomes approximately a plane wave. The fabricated antenna arrays samples have 500x500 elements, which is close to an infinite array. Therefore, the antenna arrays with different dielectric spacer layer thicknesses have approximately the same radiation patterns and effective apertures.

Fig. S1 shows the simulation results (using CST Microwave Studio) for the effective mode volume of an infinite antenna array as a function of various dielectric spacer thicknesses. Even though there is a slight increase in the mode volume as the spacer thicknesses increase (due to the increase in the energy stored in the spacer layer), the mode volume varies slowly over a large range of dielectric spacer thicknesses (20nm to 150nm) which is expected since the majority of the energy is stored in the antenna gap spacing.

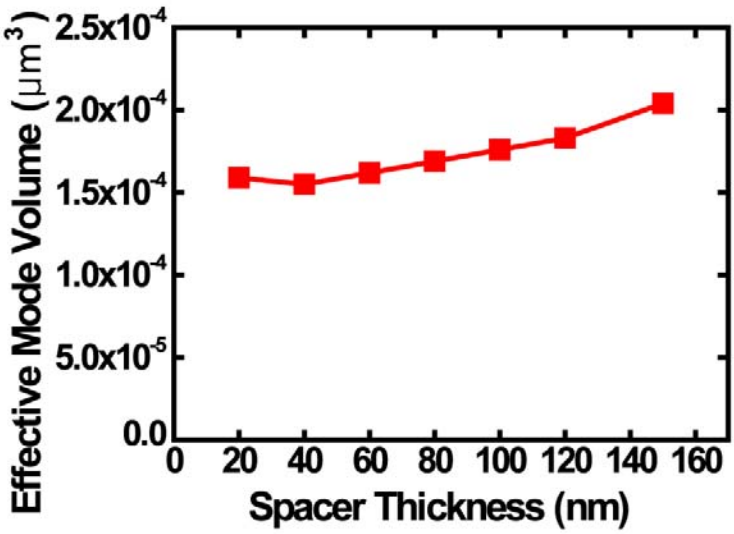

Figure S1 Effective mode volume simulations of infinite antenna arrays as a function of dielectric spacer thickness. Gold dipole antenna arrays on gold ground plane were simulated using a time domain solver based software (CST Microwave Studio). Periodic boundary condition was used to calculate an antenna array with 600 nm pitch. The plotted mode volume is calculated for one antenna element in the array.

Section IV: Characterization of the gold smoothness using a germanium adhesion layer

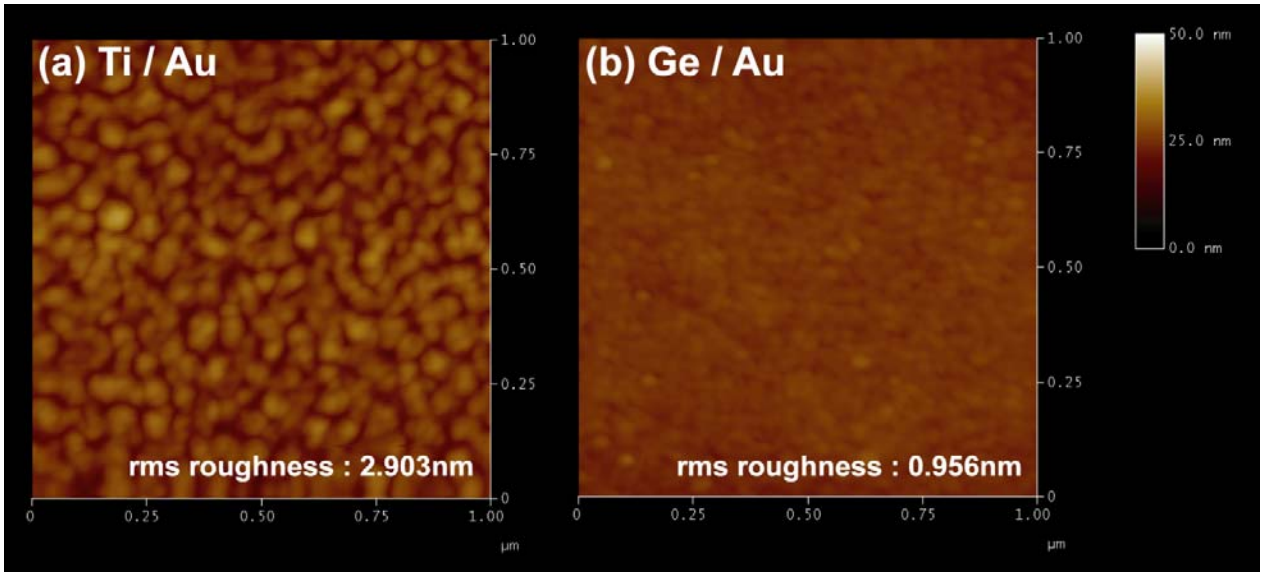

Figure S2 Comparison of the gold smoothness for titanium versus germanium adhesion layers. a, AFM measurement of 3 nm Ti and 30 nm Au evaporation on Si substrate. **b,** AFM measurement of 3 nm Ge and 30 nm Au evaporation on Si substrate. Ge adhesion layer reduces the root-mean-square (rms) surface roughness of the Au layer by 3x compared to Ti adhesion layer.

Section V: Extraction of the antenna parameters from reflection measurements

Only a certain portion of the plane wave excitation can couple to the optical antenna array, therefore, we will use the mode that is spatially matched to the antenna radiation pattern, denoted by s_{+rad}, as the input port of our CMT framework. The other part of excitation is denoted by $s_{+uncoupled}$, and the modes through

the ports s_{+rad} and $s_{+uncoupled}$ are orthogonal since they are not interacting each other. Due to the orthogonality between these modes, the total incident power($|s_+|^2$) is the sum of the powers from each mode ($|s_+|^2 = |s_{+rad}|^2 + |s_{+uncoupled}|^2$) and the relation between spatially matched input s_{+rad}, and total excitation s_+ is given by $|s_{+rad}|^2 / |s_+|^2 = A_c / A_i$ where A_c and A_i are the effective aperture of the antenna and the area of the excitation beam, respectively. The uncoupled mode ($s_{+uncoupled}$) is assumed to be reflected from the ground plane of the system ($s_{-uncoupled} = -s_{+uncoupled}$). It has been already shown in CMT theory that reflected wave can be expressed as[1]:

$$s_{-rad} = -s_{+rad} + \sqrt{\frac{1}{\tau_{rad}}}a = -s_{+rad} + \frac{\frac{1}{\tau_{rad}}}{j(\omega-\omega_0) + \frac{1}{2}\left(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{abs}}\right)} s_{+rad} \tag{S8}$$

where a is the mode amplitude in the cavity and $1/\tau_{rad}$ and $1/\tau_{abs}$ are the rates of decay due to antenna radiation and absorption, respectively. These modes s_{-rad} and $s_{-uncoupled}$ of reflected waves are also orthogonal due to the orthogonality between s_{+rad} and $s_{+uncoupled}$, and the total reflected power becomes the sum of the powers from each reflected mode. Then, we can calculate the reflectance as:

$$\begin{aligned} R(\omega) &= \frac{|s_{-uncoupled}|^2 + |s_{-rad}|^2}{|s_+|^2} = 1 - \frac{A_c}{A_i} + \frac{A_c}{A_i}\left| -1 + \frac{\frac{1}{\tau_{rad}}}{j(\omega-\omega_0) + \frac{1}{2}\left(\frac{1}{\tau_{rad}} + \frac{1}{\tau_{abs}}\right)} \right|^2 \\ &= 1 - \frac{A_c}{A_i} + \frac{A_c}{A_i}\left| -1 + \frac{\frac{\omega}{Q_{rad}}}{j(\omega-\omega_0) + \frac{1}{2}\frac{\omega}{Q}} \right|^2 \end{aligned} \tag{S9}$$

At resonance ($\omega = \omega_0$),

$$R(\omega_0) = 1 - \frac{A_c}{A_i} + \frac{A_c}{A_i}\left| -1 + \frac{2Q}{Q_{rad}} \right|^2 \tag{S10}$$

$$\therefore 1 - R(\omega_0) = 4\frac{A_c}{A_i}\frac{Q}{Q_{rad}}\left(1 - \frac{Q}{Q_{rad}}\right) \tag{S11}$$

The magnitude of reflectance dip (1-R) is a quadratic equation of Q_{rad}. This quadratic equation has a maximum peak at $Q_{rad} = 2Q$ and the maximum peak value is A_c/A_i. This condition of maximum peak is

exactly identical to the Q matching condition for the maximum field enhancement ($Q_{rad} = Q_{abs}$). With known parameters Q, R, and A_c/A_i, we can extract Q_{rad} from this quadratic equation. In fact, two solutions of the quadratic equation represent Q_{rad} and Q_{abs} due to mathematical symmetry of these two quality factors. The larger solution is Q_{rad} for the undercoupled case, and the smaller solution is Q_{rad} for the overcoupled case. Total quality factor Q and reflectance R can be extracted from a Lorentzian fit of the measured reflectance spectrum. Coupling ratio A_c/A_i cannot reach 1 since there exists spatial mismatch between excitation light and antenna radiation pattern due to finite size of antenna array and fabrication variations from individual antennas. We assume that A_c/A_i has the value of 0.65 from the maximum peak of **Fig. 4b**.

Using the equation (S1) in the supplementary section I, we find that

$$\begin{aligned} 1-R &= 4\frac{A_c}{A_i}\frac{Q}{Q_{rad}}\left(1-\frac{Q}{Q_{rad}}\right) \\ &= 4\frac{A_c}{A_i}\frac{Q}{Q_{rad}}\frac{Q}{Q_{abs}} \\ &= \frac{2\pi V_{eff}}{A_i\lambda_{res}Q_{abs}}\frac{\left|E_{loc}\right|^2}{\left|E_i\right|^2} \end{aligned} \tag{S12}$$

Therefore, the magnitude of reflectance dip is linearly proportional to field intensity enhancement. Consequently, we can assume that the field enhancement is also maximized when the size of the reflectance dip is maximized.

References:

1. Haus, H. Waves and fields in optoelectronics. *PRENTICE-HALL, INC., ENGLEWOOD CLIFFS, NJ 07632, USA, 1984, 402* (1984).
2. Maier, S.A. Plasmonic field enhancement and SERS in the effective mode volume picture. *Opt. Express* **14**, 1957-1964 (2006).
3. Stutzman, W.L. & Thiele, G.A. *Antenna theory and design*. (John Wiley & Sons, Inc.: 1998).