Design and Analysis of a Ag Rhombus Nanoparticle Film-Coupled Plasmonic Nanostructure

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ABSTRACT: We design a coupled plasmonic nanostructure, which consists of a Ag rhombus nanoparticle positioned over a silver film, separated by a dielectric spacer layer, and perform numerical analysis by calculating the radiation loss resistance of this nanostructure as the perfect electric conductor metal based on the theory of transmission line modes. Compared with the nanocube or triangular nanodisk film-coupled plasmonic nanostructures introduced in the previous works, a stronger electric field enhancement was achieved in the Ag rhombus nanoparticle film-coupled nanostructure because of the fact that the sharp tip of the rhombus nanoparticle can generate field enhancement at a hot spot. In order to demonstrate that the sharp tip can confine the electromagnetic energies strongly, we also have calculated the Purcell factor and the far-field directivity of the quantum emitter in the vicinity of this nanostructure.

1. INTRODUCTION

Plasmonic nanostructure can be taken as an optical nano-antenna; it can efficiently convert far-field radiation into a localized field and vice versa, which is because noble-metal nanostructures have a good response for plasmons. On the surface of noble-metal nanostructures (such as Au and Ag nanoparticles), electron-gas has collective oscillation properties known as the localized surface plasmon resonances. Based on this theory, many significant applications have been developed in recent years, such as optical interaction between dye molecules or quantum emitters (QEs), surface-enhanced Raman scattering, plasmon-enhanced fluorescence and luminescence, strong QE-plasmon coupling, and plasmonic laser.

Since Purcell’s work has shown that the decay rate of an excited atomic state is not only a function of the atom but also of its electromagnetic environment, a variety of plasmonic nanocavities or nanoantennas have been designed to promote the radiation of QEs or molecules, such as gold nanosphere, noble-metals nanoshell dimes, metallic nano-hole arrays, or other nanostructures. Recently, advances have shown that film-coupled nanoparticle systems (a metallic nanoparticle positioned over a metal film offers great advantages as a highly controllable system relevant for enhancing the electric field and modifying the decay rates of nearby QEs) such as the film-coupled plasmonic nanostructures of triangular nanodisks and nanocubes, also known as the metal-insulator-metal structures are applied in promoting the Purcell factor of the QE. The advantage of this film-coupled structure is that the coupling strength can be readily tuned by controlling the thickness of the dielectric spacer. Some authors have studied the electric field enhancement of the film-coupled nanostructure by means of the transmission line mode and a plasmonic circuit theory. However, the plasmonic circuit theory would become invalid when the nanoparticles have strong enough edge effects. These edge effects stem from the fact that the sharp angles of a rhombus nanoparticle funnel most of the resonant energy into the tip resulting in a larger field enhancement at the hot spot in the nanostructure. In contrast to the traditional film-coupled nanostructure including a nanocube or triangular nanodisk, in this article, we will put forward a film-coupled plasmonic nanostructure containing an Ag rhombus nanoparticle positioned over a silver film, separated by the SiO₂ spacer layer (see Figure 1), so as to obtain a stronger electric field enhancement; as for the Ag rhombus nanoparticle film-coupled plasmonic nanostructure, the side length of the rhombus is 100 nm, the height $W = 80$ nm, the thickness of the SiO₂ spacer layer $h = 10$ nm, and the Ag film is 100 nm in (b).

Figure 1. (a) Schematic diagram of the Ag rhombus nanoparticle film-coupled plasmonic nanostructure, $\alpha$ is the sharp angle of the Ag rhombus nanoparticle, the side length of the rhombus is $A = 100$ nm, the height $W = 80$ nm, the thickness of the SiO₂ spacer layer $h = 10$ nm, and the Ag film is 100 nm in (b).

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coupled nanostructure, its electric field enhancement being
used to promote the radiation of the QEs have not been
researched up to now, which will be clarified below.

The fundamental resonance of the film-coupled planar
nanoparticle arises from the transmission line mode localized
between the nanoparticle and film,\textsuperscript{29,30} where the plasmonic
resonance mode is similar to that of the microstrip antenna. To

Figure 2. (a) Extinction efficiency of the Ag rhombus nanoparticle film-coupled structures. (b) Electric field enhancement as the sharp angle $\alpha = 60^\circ$. (c) Electric field enhancement as the sharp angle $\alpha = 45^\circ$. (d) Electric field enhancement as the sharp angle $\alpha = 30^\circ$.

Figure 3. (a) Schematic diagram of the film-coupled nanostructure, the red arrow inside the SiO$_2$ film represents the QE whose orientation is vertical to the surface of the Ag substrate, $l$ denotes the distance between the QE and the center of the film. Panel (b) shows curve graphs of the QE’s Purcell factor vs wavelength, for a fixed $l = 80$ nm. The red solid line corresponds to the case of $\alpha = 30^\circ$, the green dotted line corresponds to the case of $\alpha = 45^\circ$, and the blue dotted line corresponds to the case of $\alpha = 60^\circ$. (c) Curve graphs of the QE’s Purcell factor vs wavelength, for the same sharp angle $\alpha = 30^\circ$. The red solid line corresponds to the case of $l = 70$ nm, the green dotted line corresponds to the case of $l = 80$ nm, and the blue dotted line corresponds to the case of $l = 90$ nm. (d) Directivity cross section diagram of the XZ far field of the QE in the nanostructure, where $l = 80$ nm, $\lambda = 1051$ nm, the red solid line denotes the far field directivity and radiation intensity for $\alpha = 30^\circ$, the blue dotted line corresponds to the case of $\alpha = 45^\circ$, and the black dotted line corresponds to the case of $\alpha = 60^\circ$. 
demonstrate this advantage of our design, we assume that the silver film and the Ag rhombus nanoparticle are PEC metals (for a plasmonic nanostructure, the skin depth of subwavelength electromagnetic waves in a metal cannot be ignored) and apply the transmission line mode to calculate the radiation loss resistance of the nanorhombus film-coupled structure, then analyze the electric field enhancement of this nanostructure by means of the three-dimensional finite-difference-time-domain methods (FDTD solutions v8.19, Lumerical).

2. RESULTS AND DISCUSSION

2.1. Electric Field Enhancement. The extinction efficiency of the Ag rhombus nanoparticle film-coupled structures are shown in Figure 2a the maximal values of the sharp peaks are at 957, 985, and 1051 nm when the sharp angle of the Ag rhombus nanoparticle is set as $\alpha = 60^\circ$, $45^\circ$, $30^\circ$ respectively; this peaks are the surface plasmon resonance of this nanostructure. The simulation results of electric field enhancement at the hot spot are presented in Figure 2b–d. The incident light, with the polarization direction being parallel to the long axis of the Ag rhombus, illuminates vertically on the nanostructure. Figure 2b shows that, as the sharp angle $\alpha = 60^\circ$, the maximum value of the electric field enhancement occurs at the resonance wavelength of $\lambda = 957$ nm; Figure 2c shows that, as $\alpha = 45^\circ$, the maximum field enhancement occurs at the resonance wavelength of $\lambda = 985$ nm; Figure 2d shows that, as $\alpha = 30^\circ$, the maximum field enhancement occurs at the resonance wavelength of $\lambda = 1051$ nm.

In particular, Figure 2d shows that the maximum field enhancement attains $|E_{\text{hot}}|/|E_0| > 250$ at the hot spot, which is larger than the result of 211 via a single triangular nanodisk.19

2.2. Purcell Factor and far Field Directivity. To calculate the Purcell factor of QE in the nanostructure shown in Figure 3a, we present a simulated analysis shown by Figure 3b.c. In Figure 3a, there are three film-coupled nanostructures excited by the incident light at wavelength 957, 985, and 1051 nm, respectively. As shown in Figure 3b, the smaller the sharp angle is, larger is the Purcell factor at the hot spot, which results in red-shifting of the resonance wavelength. Moreover, putting the QE into the dielectric spacer of the nanostructure with the sharp angle of $30^\circ$, which is due to the fact that the sharp angle can confine the electric field at the hot spot more strongly, and then enhance the QE’s Purcell factor more effectively. Moreover, we calculate the far field directivity and the radiation intensity of the QE and arrive at a conclusion that, as the sharp angle becomes smaller, the radiation intensity becomes stronger.

3. CONCLUSIONS

We have made an improvement for the nanocube or triangular nanodisk film-coupled structure by using the nanorhombus, so as to obtain a stronger electric field enhancement and a larger QE Purcell factor at the hot spot. Applying the theory of transmission line mode, we calculate the radiation loss resistance of the triangular nanodisk and show that the radiated loss resistance of the triangular structure is bigger than that of the nanocube, which is due to the fact that the sharp angle can confine the electric field at the hot spot more strongly, and then enhance the QE’s Purcell factor more effectively. Moreover, we calculate the far field directivity and the radiation intensity of the QE and arrive at a conclusion that, as the sharp angle becomes smaller, the radiation intensity becomes stronger.

4. COMPUTATIONAL METHODS

4.1. Radiation Loss Resistance. In our Ag rhombus nanoparticle film-coupled structure, the Ag rhombus should be regarded as consisting of two equilateral triangular nanodisks because of symmetry; our rhombus model can be simplified in terms of an equilateral triangle microstrip antenna shown in Figure 4, thereby what we need to calculate is just the radiation loss resistance of a single PEC triangular microstrip film-coupled antenna.

As shown in Figure 4, the incident light induces a resonant mode, whose electric field is vertical to the metal patch. In terms of the radiation loss mode caused by the loss magnetic modes $H_{x,y}$,29 the radiation loss power of the film-coupled antenna can be written as $P_{\text{rad}} = \int (E_x \times H_y) \cdot n dS/2U_{\text{norm}}$, where $E_x$ is the lossless electric mode, $U_{\text{norm}}$ is the normalization constant for the electric and magnetic energy in the dielectric spacer. The fictitious magnetic currents $M_{x,y}$ at the slots that

| Table 1. Summary of Calculation on Several Nanoparticle Film-Coupled Nanostructures |
|-----------------|-----------------|-----------------|-----------------|
| nanostucture    | gap size (nm)   | field enhancement | Purcell factor  |
| Ag nano rhombus | 10              | ~260             | ~5 \times 10^4 |
| Au disk array   | 25              | 100              |                |
| Au dipole antenna | 15            | ~89              |                |
| Au rod-on-mirror | 5              | ~100             | ~10^3          |
| Ag shifted cubes | 10             | 5350             |                |
| Ag nanocube     | 8               | ~100             | 10^3           |
| Au sphere-on-mirror | 1          | ~141             | 2 \times 10^6  |
| Ag triangular   | 10              | 211              |                |
| nanodisk        |                 |                  |                |

Figure 4. Schematic diagram of the equilateral PEC triangle microstrip antenna. In (a), the arrows in the dielectric spacer indicate the strengths and directions of the electric fields, the antenna is working at the TM_{001} resonant mode. In (b), the fictitious magnetic currents flow along the slot, meanwhile the directions of the electric fields are denoted by dots and crosses.
are opening along the periphery of the patch are shown in Figure 4b, which is
\[
M_x = \begin{cases} 
-2n \times E_x, & x \in [-A/2, A/2] \\
0, & \text{else}
\end{cases}
\]
where the unit vector, \( n \) is vertical to the surface of the slots shown in Figure 4, and \( E_x \) is the electric field at the surface of the slots.

For convenience, we will just consider the case that the antenna is working at the TM_{010} resonant mode. According to eq 1, in Figure 4b, the fictitious magnetic currents parallel to the \( x \)-axis have vanished along the nonradiating slot, whereas the slots adhering to the hypotenuses of the triangle microstrip antenna are the radiating slots. In the spherical coordinate frame \( (r, \theta, \phi) \), one can show the horizontal component of the electric field \( E_{\phi} = 0 \), and then the electric and magnetic fields of a single radiating slot at the far field point, \( r \) are, respectively
\[
E_{\phi} = -\frac{ik \exp(-ikr)}{4\pi r} L_{\phi}, \quad H_{\phi} = E_{\phi} \sqrt{\varepsilon_0/\mu_0}
\]
where, in terms of the y-component of the fictitious magnetic current, one has
\[
L_{\phi} = \int_S (M_{y \phi} \cos \phi) \exp(ikr \cos \phi) ds
= \int_0^{A/2} \int_0^b M_{y \phi} \cos \phi \exp[ik(y \sin \theta \sin \phi + z \cos \theta)] dy dz
= (3AE_{0h}/2) \cos \phi [\exp(Y) - 1][\exp(Z) - 1]/YZ
\]
where \( Y = i\sqrt{3}/2Ak \sin \theta \sin \phi \), \( Z = ihk \cos \theta \), \( k \) is the incident wavenumber, \( A \) is the side length of the triangular \( h \) is the thickness of the spacer layer (see Figure 4a), and \( E_0 \) is the incident light normalized value.

Now, using eq 2 the superposition of the electric fields of the two radiating slots as follows
\[
E_{\phi} = -\frac{3AkE_0 \exp(-ikr)f(\theta, \phi)}{8\pi r}
\]
where \( f(\theta, \phi) \) is a dimensionless function of the angular coordinates, and \( hkk \ll 1 \), such that \( Z = hkk \cos \theta \to 0 \), and then
\[
f(\theta, \phi) = 2 \cos(kL_e \sin \theta \cos \phi/2) \cos \phi [\exp(Y) - 1]/Y
\]
where \( L_e \) represents the effective distance between two radiating slots. Using eqs 4 and 5, the total radiated power can be written as
\[
P_{\text{rad}} = \frac{1}{2} \Re(E_{\phi}H^{*}_{\phi})
= \frac{1}{2\eta} \left[ \frac{3A}{2\lambda} \right] \left( hE_0 \right)^2 \int_0^\pi \int_0^{2\pi} f^2(\theta, \phi) \sin \theta d\theta d\phi
\]
Let \( V_0 = hE_0 \eta = \sqrt{\mu_0/\varepsilon_0} \), it follows from eq 6 that
\[
P_{\text{rad}} = \frac{3V_0^2}{8\pi} \left( \frac{L}{\pi^2} \right) = \frac{V_0^2}{R_{\text{loss}}}
\]
where \( R_{\text{loss}} \) represents the radiation loss resistance of the PEC patch antenna, and
\[
R_{\text{loss}} = 8\pi^2/3L
\]
\[
I_1 = \int_0^\pi \int_0^{2\pi} \frac{\cos^2 \phi [\exp(Y) - 1]^2}{\sin \theta \sin^2 \phi} \cos^2(kL_e \sin \theta \cos \phi/2) d\theta d\phi
\]
Now, using eq 8, we will calculate the radiation loss resistance of the triangular nanodisk given by Figure 4a, with the result shown via Figure 5a furthermore, the far field scattering cross section of the triangular nanodisk and the nanocube are simulated by the FDTD solutions shown in Figure 5b (note: these nanostructures as the plasmonic nanoantenna are no longer the PEC patch antenna). For comparing our results with the previous results provided by reference 23, the side length and thickness of our triangular nanodisk are taken as the same as the ones of the nanocube in ref 23.

The total power can be written as \( P_{\text{total}} = P_{\text{cav}} + P_{\text{rad}} \), where \( P_{\text{cav}} \) denotes that the power is confined to the nanostructure enhancing the electric field, whereas the radiated power, \( P_{\text{rad}} \) is launched by the radiating slots. For a given total power, the larger \( P_{\text{cav}} \) is, the smaller \( P_{\text{rad}} \) is, and vice versa. On the other hand, eq 7 implies that the larger \( R_{\text{loss}} \) is, the smaller \( P_{\text{rad}} \) is. As a result, Figure 5 shows that, compared with the nanocube, the radiating slots in the triangular microstrip antenna have a smaller radiated power, thereby most of the EM energy is confined to the rhombus nanoparticle film-coupled nanostructure.

4.2. Purcell Factor of QE. Estimating the capability of QE’s radiation by calculating the Purcell factor is the universal way, the QE emission decay rate, \( \gamma \), has been given by Fermi’s
golden rule. To calculate the QE’s emission decay rate in the dielectric space conveniently, Lukas Novotny has given the expression for the decay rate \( \gamma = \frac{n_0 e_l |p|^2 \rho_m}{3 \hbar c} \). \( |p| \) is the transition dipole moment, and \( \rho_m \) is a plasmon mode density in the dielectric space.

The electric field enhancement implies that a plasmon mode density is also promoted at the hot spot (see eq 12 later), and then the QE’s Purcell factor would be boosted. In fact, let us assume that the plasmonic nanostructure is working at the plasmonic mode with the oscillation frequency of \( \omega_{m} \) when the QE decay rate, \( \gamma'_{av} \), is enhanced relative to the free-space decay rate, \( \gamma_{av} \) and the Purcell factor is expressed as

\[
F_p = \frac{\gamma'_{av}}{\gamma_{av}} = \frac{6\pi Q_m}{k^3 V_m}
\]

where \( Q_m \) is the mode quality factor, and \( V_m \) is the mode volume in the dielectric space. For the moment, the plasmon mode density, as the position function of \( r \) in the dielectric spacer, can be defined as the inverse of \( V_m \).

\[
\rho_m(r) = \frac{1}{V_m(r)} = \frac{2E_m^2(r)}{\int dV E_m^2(r) d(\omega_m, \omega')} d\omega_m
\]

where \( E_m(r) \) is the electric field, \( V \) is the geometric volume of dielectric spacer, and \( \omega' \) is the real part of the metal Ag dielectric constant.

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### Notes

The authors declare no competing financial interest.

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