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Cavity modes and their excitations in elliptical plasmonic patch nanoantennas

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Abstract: We present experimental and theoretical studies of two dimensional periodic arrays of elliptical plasmonic patch nanoantennas. Experimental and simulation results demonstrate that the azimuthal symmetry breaking of the metal patches leads to the occurrence of even and odd resonant cavity modes and the excitation geometries dependent on their modal symmetries. We show that the cavity modes can be described by the product of radial and angular Mathieu functions with excellent agreements with both simulations and experiments. The effects of the patch periodicity on the excitation of the surface plasmon and its coupling with the cavity modes are also discussed.

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References and links
1. Introduction

Plasmonic nanoantennas have attracted significant attention in recent years due to their ability in coupling free space electromagnetic radiation into sub-diffraction limited volumes and vice versa based on the electrodynamics reciprocity [1, 2]. Resonantly enhanced local fields in these nanoantennas facilitate applications in various fields including surface enhanced fluorescence and Raman spectroscopy [3–6] and in nonlinear optics [7,8]. For examples, as the optical analogues to their radio frequency counterparts, Hertzian dipole and Yagi-Uda antennas enable not only focusing light into nanometer spots but also emitting light unidirectionally by coupling quantum dots with them [9–11]. Vertical metal-dielectric-metal (MDM) designs of plasmonic nanoantennas and nanocavities have been proposed and demonstrated with the great advantage that the nanogap thickness can be precisely controlled by using the advanced thin film deposition techniques [12–14].

A variant of the MDM nanoantennas is the plasmonic patch nanoantennas where metal patches are fabricated on a metal film with a dielectric spacer layer [15]. The large local field enhancements in these patch nanoantennas make them attractive substrates for surface enhanced Raman spectroscopy (SERS) [15–17]. Recent studies show that the plasmonic patch antennas provide a new avenue towards various applications such as near perfect absorbers [18–21], single photon light sources by coupling them with quantum emission systems [22], metamaterials [23], and biosensors [24]. By exciting the cavity modes between two metal
plates (a configuration similar to the patch antennas), a strong light-induced negative optical pressure can be introduced [25]. To facilitate the design of plasmonic patch nanoantennas for these various applications, a simple approach to accurately predict the resonant frequencies of these patch nanoantennas is highly desired. Previous studies, however, are mostly focused on the first cavity mode for the circular or square patch shapes, while the high order cavity modes and their resonant/excitation conditions have rarely been addressed quantitatively.

In this letter, we present the physical understanding of the cavity modes and their excitation conditions for plasmonic patch nanoantennas in optical frequencies. Patches with elliptical shapes were used to investigate the effect of the azimuthal symmetry breaking on the local electrical field distributions for different cavity modes and on their excitation conditions. Numerical calculations show that breaking the circular symmetry leads to the presence of both even and odd cavity modes, and that the excitation configurations for these modes are dictated by their modal symmetries. Analytical expressions of the modal field distributions agree well with the simulation results. By using the actual patch radii plus the gap thickness as the effective radii, we show that the resonant condition based on Neumann boundary conditions show in excellent agreements with experimental and simulation results. This physical understanding of the resonant modes and their excitation conditions of the patch nanoantennas should be extendable to plasmonic patch nanoantennas with other geometrical shapes.

2. Sample fabrication

To fabricate the plasmonic patch nanoantennas [Fig. 1(a)], a 10 nm NiCr adhesion layer and a 100 nm Ag film were sequentially deposited on a Si wafer using electron beam evaporation, and then a 15 nm Al$_2$O$_3$ dielectric layer was deposited using atomic layer deposition (ALD). The 45 nm thick top elliptical Ag patches were fabricated using the standard electron beam lithography and lift-off processes. The reason that we used Ag instead of Au is to design the resonant wavelengths of the first few cavity modes below 950nm (the limit of our imaging spectrograph system). The advantages of using ALD process include the conformal and uniform film coverage and sub-nm accuracy in thickness control. An exemplary SEM picture of the fabricated patch nanoantennas is shown in Fig. 1(b). To measure the reflection spectra, a collimated white light beam was focused onto the samples using a 40 × (0.6NA) objective, and the reflected light was collected using the same objective for spectral measurements. The reflection spectra are normalized by the spectra measured from the areas without the patches.

![Fig. 1. (a) Schematic elliptical plasmonic patch nanoantennas. The sky blue layers represent Ag, and the dark blue represents Al$_2$O$_3$. Grey is the silicon substrate. (b) An SEM image of a 2D array of plasmonic patch nanoantennas with a period of 500nm and two major axis radii a = 72nm, b = 52nm. The long axis of the patches is tilted about 22° from vertical direction.](image)

The major axial radii of the elliptical patches are varied from 40 nm to 96 nm. Two patch periods, 300 nm and 500 nm, are used for the patch antenna samples. As will be discussed later, these patch periods are small enough to ensure minimal effect of surface plasmons excitation outside the cavity on the cavity modes, and large enough to ensure no overlapping of the fringe fields between neighboring patches.

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3. Excitation of the cavity modes: experiments and simulations

The measured reflection spectra for two representative patch sizes are presented in Fig. 2(a) and 2(c), where several dips with different absorption depths can be observed. The resonant wavelengths of these dips shift to blue when the incident polarization is changed from the direction parallel to the long axis to the direction parallel to the short axis of the elliptical patches.

To find the physical origins of these resonant dips, we performed numerical calculations using the finite integration technique (CST-Microwave Studio). The frequency dependent permittivity for silver was described by the Drude model $\varepsilon_{Ag} = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega(\omega + i\gamma)}$, with $\varepsilon_{\infty} = 3.57$, $\omega_p = 1.388 \times 10^{16}$ rad/s, and $\gamma = 1.064 \times 10^{14}$ Hz obtained by fitting to the measured bulk permittivity of Ag [26]. The optical constants from both Ref [26] and [27] have been used in numerical simulations in the previous studies such as in [28]. Since the imaginary part of the permittivity from Ref [26] is smaller than that from Palik’s Handbook, simulations normally result in narrower resonances. While the real part of the permittivity for Ag from these two sources differs by only a small percentage for wavelength below 1 µm, the difference in the predicted resonant frequencies is normally very small as verified in also our simulations. The dielectric permittivity of $\text{Al}_2\text{O}_3$ was fixed at 2.40. To emulate the experimental conditions, the reflection spectra were obtained by averaging over two incident angles (0° and 20°) for two polarizations parallel to the major axes of the patches respectively. It can be observed that the calculated reflection spectra reproduce these resonant features in experimental data at similar resonant wavelengths [Fig. 2(a-d)]. At these resonant wavelengths, the calculated near-field spectra exhibit resonant peaks, indicating cavity resonances [dotted lines in Fig. 2(b) and 2(d)].

We calculated the $z$-component of the electrical field distributions in the $x$-$y$ plane through the middle of the $\text{Al}_2\text{O}_3$ layer at these resonant wavelengths. Figure 3 presents the results for different cavity modes and their corresponding excitation conditions. Based on comparing
these simulated field distribution patterns with the theoretical results in next section, these cavity modes and their presences in the far-field spectra have be indexed (Fig. 2 and 3). The excitations of these cavity modes are sensitive to both incidence angle and polarization directions of the exciting light and will be discussed later in this paper.

4. Discussion

The cavity modes observed in the elliptical patch nanoantennas are due to constructive interferences of gap surface plasmons generated and reflected at the patch boundaries propagating parallel to the interface. To understand these cavity modes, we consider the electromagnetic waves propagating in the metal-dielectric-metal (MDM) waveguide structure. The electrical field $E$ can be described by the Helmholtz Eq. $\nabla^2 E + k_i^2 E = 0$ where $k_i^2 = \varepsilon_i \omega^2 / c^2$ are the total wave vectors with the subscript $i = m, d$ referring to metal and dielectric regions, $\omega$ and $c$ are the radial frequency and speed of light in vacuum. Since the gap surface plasmons need to be evanescent in the direction (z) perpendicular to the MDM plane, the propagation constant along $z$ should be imaginary, denoted as $ik_{gsp}$. Therefore, the total wave vector $k_i$ is the summation of the propagation constant $ik_{gsp}$ and the propagation constant along the MDM plane or the gap plasmon wave vector $k_{gsp}$, $k_i^2 = k_{gsp}^2 - k_{gsp}^2$.

Fig. 3. Excitation configurations and the snapshots of simulated electrical field ($E_z$) distributions for different cavity modes for the 93nm × 74nm patches: (a) modes symmetrical to x-axis and anti-symmetric to y-axis; (b) modes symmetrical to y-axis and anti-symmetric to x-axis; (c) modes symmetric to both x- and y-axes; (d) modes anti-symmetric to both x- and y-axes.

There exist two gap surface plasmon modes with one symmetric and one anti-symmetric profile of field distribution [29]. For small dielectric thicknesses, the symmetric mode exists at high frequencies and can be ignored in our case. For the top metal layer of the MDM structure with a finite thickness $t_m$, the dispersion relation for the anti-symmetric mode can be expressed as [30]:

$$k_{gm}^2 \varepsilon_m = \frac{-1 + \sqrt{r_1 + r_2 + r_1 r_2}}{1 + \sqrt{r_1 + r_2 + r_1 r_2}},$$

where $k_{gm} = \sqrt{k_{gsp}^2 - \varepsilon_m \omega^2 / c^2}$, $k_{gd} = \sqrt{k_{gsp}^2 - \varepsilon_d \omega^2 / c^2}$, $r_1 = \exp(-2k_{gm} t_m)$, $r_2 = \exp(-2k_{gd} t_d)$, and $t_d$ is the thicknesses of the dielectric layer.
Given the elliptical shape of the nanocavities, it is convenient to consider the Helmholtz Eqs. in the elliptical cylindrical coordinates \((\xi, \eta, z)\). The transformation from the Cartesian coordinates \((x, y, z)\) to the elliptical coordinates is:

\[
\begin{align*}
\xi &= f \cosh \zeta \cos \eta, \\
y &= f \sinh \zeta \sin \eta, \\
z &= z
\end{align*}
\]

where \(f\) is the focal length of the ellipse, \(f = (a^2 - b^2)^{1/2}\). Since our primary interest is in the electrical field distributions inside the dielectric layer, we focus only on the dominant component of the electrical field \(E_{zd}(\xi, \eta, z)\). After the transformation, the Helmholtz Eq. for \(E_{zd}(\xi, \eta, z)\) can be written as [28,29]:

\[
\frac{1}{f^2 (\sinh^2 \xi + \sin^2 \eta)} \left( \frac{d^2 E_{zd}}{d\xi^2} + \frac{d^2 E_{zd}}{d\eta^2} \right) + \frac{d^2 E_{zd}}{dz^2} + k^2 E_{zd} = 0, \quad (2)
\]

Assuming \(E_{zd}(\xi, \eta, z) = R(\xi)\Phi(\eta)Z(z)\), separation of variables decomposes Eq. (2) into the radial and angular Mathieu Eqs [25,30]:

\[
\begin{align*}
\frac{d^2 R}{d\xi^2} + (c - 2q \cosh 2\xi)R &= 0, \\
\frac{d^2 \Phi}{d\eta^2} + (c - 2q \cos 2\eta)\Phi &= 0,
\end{align*} \quad (3)
\]

where \(c\) is the separation constant and \(q = f^2 k^2_{sp}/4\). The fact that \(\Phi\) should be a periodic function of \(\eta\) with either a \(\pi\) or \(2\pi\) period determines the possible eigen values of \(c\) [31,32]. The allowed solutions for \(\Phi\) are two independent families of even \((e)\) and odd \((o)\) angular Mathieu functions: \(Ce_m(\eta, q)\) for \(m \geq 0\) and \(So_m(\eta, q)\) for \(m \geq 1\) respectively. The radial function \(R\) should be a non-periodic, decreasing oscillatory function of \(\xi\) in \(0 \leq \xi < \infty\) with non-singularity at the origin; the standing wave solutions for \(R\) are the even and odd radial Mathieu functions of the first kind: \(Je_m(\xi, q)\) for \(m \geq 0\) and \(Jo_m(\xi, q)\) for \(m \geq 1\). For fixed \(z\), \(E_{zd}(\xi, \eta, z)\) is proportional to the product of the angular and radial Mathieu function:

\[
E_m^{\alpha} \approx \begin{cases} 
Je_m(\xi_0, q)Ce_m(\eta, q), m \geq 0 \text{ (even)} \\
Jo_m(\xi_0, q)Se_m(\eta, q), m \geq 1 \text{ (odd)}
\end{cases}, \quad (4)
\]

where the superscript \(m\) stands for the \(m\)-th order. Since the nanocavities have an open edge, the electrical field \(E_{zd}(\xi, \eta, z)\) is approximately at its local maximum at the boundary. As a result, the cavity resonances are determined by the Neumann boundary condition:

\[
\begin{align*}
\text{(even)} : & \ J'e_m(\xi_0, q)Ce_m(\eta, q) = 0, \\
\text{(odd)} : & \ J'o_m(\xi_0, q)Se_m(\eta, q) = 0, \quad (5)
\end{align*}
\]

where \(\xi_0 = \arcsin h(b/f)\) defines the cavity boundary. For a given order \(m\), there exists an infinite number of \(q\) values satisfying Eq. (5). We use \(q_m^{\alpha}\) to denote the \(n\)-th zero of \(J'e_m(\xi_0, q)\) or \(J'o_m(\xi_0, q)\), and use \(e_m\) and \(a_m\) to denote the even and odd cavity modes.
Based on Eq. (4) and Eq. (5), we calculated the electrical field distributions for these cavity modes of lowest orders. The results as shown in Fig. 4 are in excellent agreements with the field distributions obtained in simulations (Fig. 3). Previous studies of elliptical patch antennas in microwave frequencies for metamaterials show similar field distribution patterns though with detailed structures are different from ours [23].

As can be seen in Fig. 3, the excitation conditions for these cavity modes are dictated by their modal symmetries. The cavity modes with odd m can be excited with both normal and tilted illumination, while the excitation configurations for the even and odd modes are rotated by 90° from each other. While for the even modes ($e_{21}$, $o_{02}$) with even m which are symmetric to both x- and y-axes, tilted illumination with p-polarization is required [Fig. 3(c)]; and for the odd modes with even m which are anti-symmetric to both x- and y-axes ($o_{21}$), tilted illumination with s-polarization is needed. As a result, these modes excited only by tilted illumination have small resonant signatures in the far-field reflection spectra (Fig. 2).

These cavity excitation rules are set by the phase asymmetry of the excited gap surface plasmons. For normal incidence, the electric fields $E_{zd}$ at the opposite cavity sides are always in opposite phase along the polarization direction while in the same phase along the direction perpendicular to the polarization [Fig. 3(a-b)]. Consequently, in order to excite cavity modes such as $e_{21}$, $e_{02}$ or $o_{02}$ this asymmetry has to be broken by using tilted illumination. It is important to note that due to the subwavelength size of the patches, the tilted illumination cannot completely compensate the $\pi$ phase difference of the gap plasmons excited at the opposite edges, and the modal field distributions inside the cavities are always composed of a standing wave superposed on a propagating wave. This also makes it difficult to identify cavity modes such as $e_{03}$, because their appearances in the far-field spectra are expected to be shallow [red curves in Fig. 2(a, b)].

For one given $q_{mn}^{\alpha\beta}$, the gap surface plasmons forming the standing waves should satisfy:

$$k_{gap} = (4q_{mn}^{\alpha\beta} / f^2)^{1/2},$$

To validate this resonant condition, we calculated the $k_{gap}$ for these cavity modes by using Eqs. (5-6) based on their resonant wavelengths and patch sizes observed in the experiments, and compare it with the $k_{gap}$ calculated using the dispersion relation (Eq. (1)). The results for different patch sizes in Fig. 5(a) show reasonable agreements between the resonant condition and the gap plasmon dispersion curve. To note here, we only included the data for $e_{11}$/$o_{11}$, $e_{12}$/$o_{12}$ and $e_{31}$/$o_{31}$ modes in Fig. 5 because their features can be easily identified in the measured reflection spectra.
Fig. 5. Data points represent the measured cavity resonant frequencies versus the gap plasmon wave vector calculated using Eq. (6) for real patch sizes (a) and for the effective patch sizes $a' = a + h$, $b' = b + h$ (b). The blue solid curves represent the dispersion curve for the gap plasmons calculated using Eq. (1).

It is also discernible that the resonant frequencies obtained using Eq. (6) are systematically lower than the dispersion curve for the gap surface plasmons obtained using Eq. (1). This systematic discrepancy can be ascribed to the fringing fields, i.e. the electrical fields do not go to zero beyond the cavity edge. In another word, the antinodes of the cavity modes are not exactly located at the cavity boundary, and there exists effectively a phase shift upon excitation/reflection of the gap surface plasmons at the boundary [12,33]. This fringing field effect can also be considered as a result of the capacitance between the patch edges and the bottom metal film [34,35]. To take this fringing field into account, one simple while effective empirical approach suggested in microwave frequencies is to use the sum of the actual patch radius and the dielectric layer thickness as the effective radius [36]. By following this empirical correction, we recalculated the resonant frequencies using $a' = a + h$ and $b' = b + h$ as the two main axial radii in Eq. (6). The results show excellent agreements with the dispersion for gap surface plasmons [Fig. 5(b)].

One interesting observation is that the resonant frequencies for even mode and odd modes are noticeably different except for the e$_{31}$ and o$_{31}$ modes. This is fortuitous and because the resonant conditions for these two modes are almost the same. For example, the value of $q_{31}$ and $q'_{31}$ are 1.9159 and 1.963 for the same patch size with $a = 93$nm, $b = 74$nm respectively.
Fig. 6. Snapshots of the z-component $E_{zd}$ of the electric field calculated at the middle plane through the dielectric layer at the resonant wavelength 540 nm for two different patch radii: 72nm × 52nm (a) and 93nm × 74nm (b). The illumination is normal to the plane.

The effects of array periods on the resonant wavelengths are very interesting while quite complicated. We speculate that there exist three regions: (1) For small periods where the fringe fields between neighboring patches overlap, the cavity modes are coupled with each other (in analog to the systems of atoms with overlapping wave functions), and form a 2D photonic crystal system. In this case, it can be expected that the gap plasmons will be delocalized at certain frequency bands, while be prohibited to propagate forming band gaps at the other frequencies. (2) For large periods, the surface plasmons at the Ag/Al$_2$O$_3$ interface outside the cavities are often excited, causing the complex interactions between cavity modes and surface plasmon modes. (3) For the intermediate periods like those in our experiments, the cavity fringe fields are not overlapping, and the surface plasmon modes are not crowded. In the following, we show numerical studies for the last two situations as a proof that the cavity modes and resonant conditions in our case are not affected by these array effects.

The 2D periodicity of the metal patches provides a reciprocal vector to compensate for the momentum mismatch between the incident light and the surface plasmon waves at the Ag-Al$_2$O$_3$ interface; a natural question is therefore how the surface plasmon excitation outside the cavity affects the cavity resonances. Although the incident angles vary in a range determined by the objective N.A., the excitation condition for the surface plasmons propagating at the direction perpendicular to the incident plane remains unchanged. For example, for 500 nm array period, the dips at about 540 nm in the reflection spectra are due to the excitation of the surface plasmons. This can be verified by looking at the local field distributions at this resonant wavelength, where the interference patterns of surface plasmons outside the cavities can clearly be seen. For a small thickness (15nm) of the Al$_2$O$_3$ film, the dispersion for the surface plasmon waves at the Ag-Al$_2$O$_3$ interface can be approximated as

$$k_{sp} = k_{sp0} + \Delta k$$

where $k_{sp0} = \frac{\omega}{c} \sqrt{\varepsilon_m'/(\varepsilon_m''+1)}$ is the wave vector for the excited surface plasmons at the Ag-air interface, $\Delta k = \frac{\omega}{c} \frac{\varepsilon_d - 1}{\varepsilon_d' - \varepsilon_m'} \frac{1}{1 - \varepsilon_m' \sqrt{\varepsilon_m''}} \frac{2\pi}{\lambda}$, $\varepsilon_m'$ is the real part of the permittivity for Ag, and $\varepsilon_d'$ is the permittivity for Al$_2$O$_3$ [37]. Simple calculations show that for the incident light at 540 nm wavelength, $k_{exp} = 0.0125$ nm$^{-1}$ matches with the reciprocal vector of the patch arrays.
Fig. 7. Calculated reflection spectra for different periods with the elliptical patch size: \(a = 70\) nm; \(b = 50\) nm. a) and c): periods vary between 300nm to 500nm; b) and d): periods vary between 600nm to 900nm. a) and b): Incident polarization is along the long axis of elliptical patches for; c) and d): Incident polarization is along the short axis of elliptical patches.

The coupling effects between surface plasmons and cavity resonances usually induce Fano type resonances. For a fixed patch period, the excited surface plasmon may couple with different cavity modes for different patch sizes. As it can be seen in Fig. 6, for 500 nm array period, the surface plasmon excited by 540 nm wavelength light can couple with either \(e_{11}\) mode or \(e_{31}\) mode depending on different patch sizes. It is also interesting to note that for the \(e_{11}\) mode not at resonance, the surface plasmon waves outside and inside the nanocavities are in opposite phase [Fig. 6(a)]. While for the \(e_{31}\) mode which is at resonance, the electrical fields due to the surface plasmons outside and inside the nanocavities are in phase [Fig. 6(b)]. It is important to note that the small patch periods (300 nm and 500 nm) are used in the experiments to minimize the excitations of surface plasmons outside the cavities and their effects on the cavity resonances. This can be seen from the simulated reflection spectra for different patch periodicities (Fig. 7). For the patch period below 500nm, the narrow dips due to the excitation of surface plasmons shift to red with the period increase, while the reflection dips due to the cavity modes remain unchanged as long as they are not overlapping with the surface plasmon excitation [Fig. 7(a)]. When the patch period is larger than 600 nm, the resonant wavelengths of these cavity modes start to vary with the changes in the patch period due to the excitation of multiple surface plasmon modes [Fig. 7(b)].

5. Conclusion

In summary, we have studied the two dimensional periodic arrays of elliptical plasmonic patch nanoantennas. It is shown that by breaking the azimuthal symmetry with elliptical patch shapes, even and odd resonant cavity modes can be excited with the excitation configurations depending on their modal symmetries. An analytical expression for the cavity modal field distributions based on Mathieu functions has been derived, yielding excellent agreements with both simulations and experiments.

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