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Dual-band plasmonic resonator based on Jerusalem cross-shaped nanoapertures

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Abstract

In this paper, we both experimentally and numerically introduce a dual-resonant metamaterial based on subwavelength Jerusalem cross-shaped apertures. We numerically investigate the physical origin of the dual-resonant behavior, originating from the constituting aperture elements, through finite difference time domain calculations. Our numerical calculations show that at the dual-resonances, the aperture system supports large and easily accessible local electromagnetic fields. In order to experimentally realize the aperture system, we utilize a high-precision and lift-off free fabrication method based on electron-beam lithography. We also introduce a fine-tuning mechanism for controlling the dual-resonant spectral response through geometrical device parameters. Finally, we show the aperture system's highly advantageous far- and near-field characteristics through numerical calculations on refractive index sensitivity. The quantitative analyses on the availability of the local fields supported by the aperture system are employed to explain the grounds behind the sensitivity of each spectral feature within the dual-resonant behavior. Possessing dual-resonances with large and accessible electromagnetic fields, Jerusalem cross-shaped apertures can be highly advantageous for wide range of applications demanding multiple spectral features with strong nearfield characteristics.

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1. Introduction

Plasmonic subwavelength systems based on metallic nanoapertures or nanoparticles have received significant

attention as they have very advantageous optical properties, which enable extraordinary physical phenomena, e.g., from metamaterials to perfect lensing [1–4]. Recently accessing excitation-forbidden dark modes, systems with extraordinary optical properties have been introduced [5,6]. Electromagnetic properties of plasmonic systems could also result in multi-resonant spectral responses derived from the collective mechanism of

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constituting resonating elements [7–9]. Optical properties of these multi-resonant systems are highly dependent on the refractive index of surrounding media [10], shape and orientation of the apertures [11,12], aperture size [12,13], metal thickness [14], and the lattice geometry of the aperture arrays [15–25]. This dependence provides a fine-tuning mechanism of their multi-resonant spectra in wide wavelength ranges. Recent advancements in computational electromagnetics and nanofabrication techniques enable numerical and experimental analyses of these nanostructures, which provide tunable multi-resonant spectral responses [26–32]. For instance, microwave dual-resonant metamaterials have been introduced for extraordinary refractive index properties [33,34], while near-infrared multi-spectral nanoapertures have been investigated for negative indexing [35,36]. In this sense recently, Jerusalem cross (JC)-shaped nanostructures have been investigated due to their extraordinary far- and nearfield characteristics [37–41].

In this paper, we experimentally and numerically investigate a dual-band resonator system based on JC-shaped aperture arrays. We show the physical origin of the dual-resonant behavior, deriving from the constituting aperture elements in the compact system, through finite difference time domain (FDTD) simulations. We also introduce a fine-tuning mechanism for controlling the dual-resonant optical response via geometrical parameters. We show that the aperture system supports large and highly accessible local electromagnetic

fields, significantly improving light-matter interaction in nanometer scale, which is highly important for surface-enhanced applications demanding strong nearfields. We also perform numerical calculations in order to quantitatively show the accessibility of local electromagnetic fields. Highly advantageous nearfield characteristics of the nanoapertures are shown through numerical calculations on refractive index sensitivity, where the system is embedded in different refractive indices of bulk solutions. Finally, we utilize a high-precision and lift-off free fabrication method based on electron beam lithography to experimentally realize JC-shaped apertures. Possessing a dual-resonant spectral response with large and accessible nearfields, JC-shaped apertures can be highly advantageous for wide range of applications including ultra-sensitive biosensing and spectroscopy, wavelength-tunable active filters, optical modulators, and ultrafast optical switching devices.

2. Dual-band JC-shaped nanoaperture system

The proposed JC-shaped aperture system is schematically illustrated in Fig. 1a. Aperture arrays are fabricated through a 50 nm gold film standing on a 100 nm free-standing silicon nitride membrane. Fig. 1b shows the scanning electron microscopy (SEM) image of the aperture system, where the device parameters are: $s = 600$ nm, $g = 470$ nm, $w = 200$ nm, $L = 1150$ nm and $P = 1400$ nm (array period). Fig. 1c and d shows the calculated and

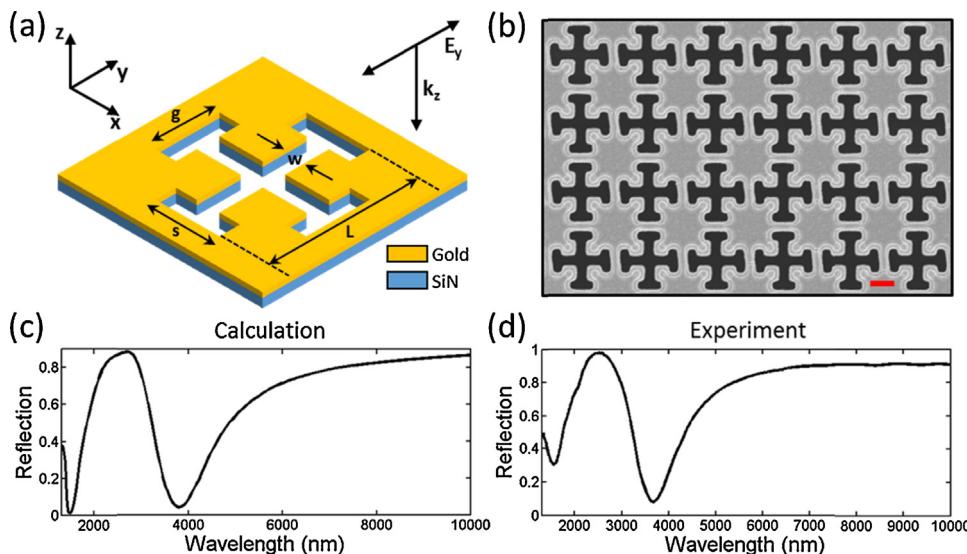


Fig. 1. (a) Schematic illustration of the JC-shaped aperture. In figure, geometrical device parameters of the aperture system along with the polarization and propagation directions of the illumination source are shown. (b) SEM image of the fabricated aperture array. Scale bar, 500 nm. (c) Numerical and (d) experimental reflection spectra of the JC-shaped aperture. Corresponding device parameters are: $s = 600$ nm, $g = 470$ nm, $w = 200$ nm, $L = 1150$ nm, and $P = 1400$ nm. Thicknesses of the silicon nitride (SiN) membrane and gold film are 100 nm and 50 nm, respectively.

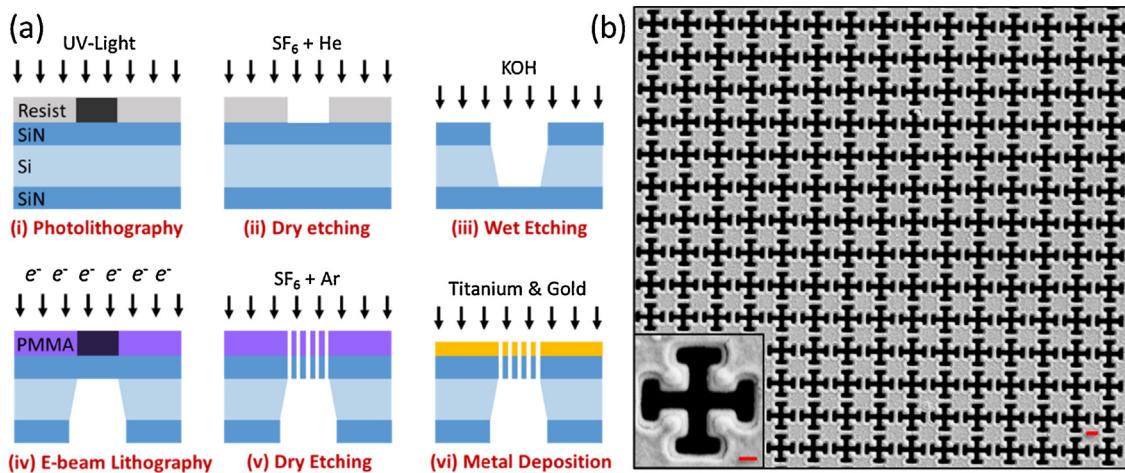


Fig. 2. (a) Steps of the lift-off free nanofabrication method. (b) SEM images of the fabricated nanoapertures. Scale bar (zoom image), 500 nm (200 nm). Corresponding device parameters are: $s = 600$ nm, $g = 470$ nm, $w = 200$ nm, $L = 1150$ nm, and $P = 1400$ nm. Thicknesses of the silicon nitride membrane and gold film are 100 nm and 50 nm, respectively.

experimental reflection spectra under a y -polarized light source (parallel to the top and bottom aperture arms), demonstrating their good correlation, where the aperture system supports two distinct resonances within the spectral region of interest. For experimental measurements, we collect the reflection data using a Fourier transform infrared (FTIR) spectrometer (Bruker, IFS 66/s) coupled to an infrared microscope (Bruker, Hyperion 1000) with a resolution of 4 cm^{-1} , which consists of 256 scans co-added with a mirror repetition rate of 40 kHz. In order to eliminate atmospheric water vapor absorption within the spectral region of interest, data collection is performed under a dry air purged environment. Reflection spectrum is determined through the ratio between the reflected light from the aperture system and the reflected light from a perfect mirror made of an optically thick gold film.

3. Fabrication of JC-shaped apertures

For experimental realization, we utilize a lift-off free nanofabrication technique based on electron-beam lithography, as schematically illustrated in Fig. 2a. We utilize a $500\text{ }\mu\text{m}$ silicon wafer with 100 nm low-pressure chemical vapor deposited (LPCVD) silicon nitride films on both sides. (i) We perform photolithography on a $2\text{ }\mu\text{m}$ positive photoresist (MICROPOSITTM S1818) using SUSS MicroTec MA/BA6 Mask Aligner. After development, we have $750\text{ }\mu\text{m} \times 750\text{ }\mu\text{m}$ square aperture patterns on the bottom silicon nitride film. (ii) We perform dry etching (Plasma-Therm Reactive Ion Etcher) using SF_6 and He gases in order to completely remove the silicon nitride film within the transferred

pattern at the back side of the wafer. (iii) Chips are then immersed in KOH solution to selectively remove the silicon substrate under the top silicon nitride film. (iv) We perform electron-beam lithography (Zeiss SUPRA 40VP) on a positive resist, polymethyl methacrylate (950 PMMA A5, MICROCHEM). (v) After development on the patterned PMMA using methyl isobutyl ketone and isopropyl alcohol (MIBK-IPA, 1:3) solution, we perform dry etching (while utilizing PMMA as a mask) with SF_6 and Ar gases in order to remove silicon nitride film, which realizes apertures through free-standing silicon nitride membranes. (vi) Finally, we deposit 5 nm titanium (adhesion layer) and 50 nm gold (CHA Electron Beam Evaporator), which realizes apertures through the gold film in a lift-off free manner. SEM images of the fabricated system in Fig. 2b demonstrate that the apertures are well-defined and smooth (zoomed image) as well as the aperture array is uniform over large areas.

4. Origin of dual-resonant response

In order to show the physical origin of the dual-resonant behavior, we perform numerical analyses through FDTD calculations. In simulations, dielectric constants of gold and titanium are taken from Ref. [42] and that of silicon nitride is experimentally obtained through ellipsometry measurements. For the unit cell consisting of a single JC-shaped aperture, periodic boundary condition is used along x - and y -directions in order to create the array behavior (aperture array is along xy -plane) and perfectly matched layer boundary condition is used along z -direction (direction of propagation). Mesh sizes are 5 nm in all directions

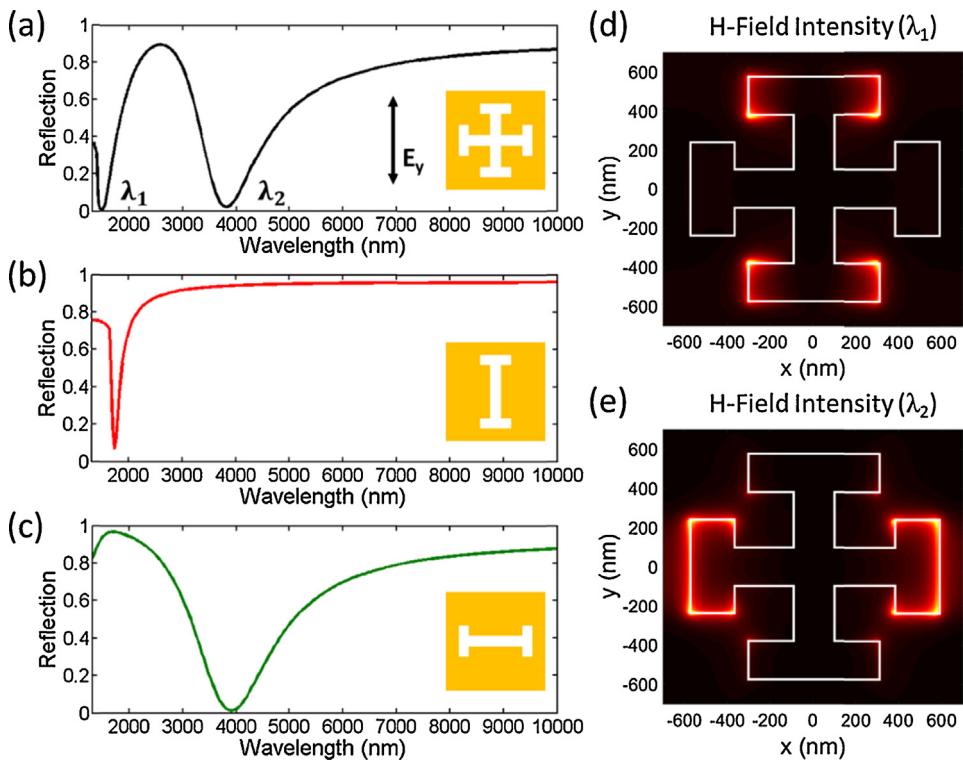


Fig. 3. Calculated reflection spectra of (a) the compact JC-shaped aperture, and the constituting (b) vertical and (c) horizontal H-shaped apertures. Figure inset shows the polarization direction of the illumination source. Magnetic field intensity ($|H|^2$) distributions for the JC-shaped aperture calculated at (d) $\lambda_1 = 1490$ nm and (e) $\lambda_2 = 3800$ nm. Corresponding device parameters are: $s = 600$ nm, $g = 470$ nm, $w = 200$ nm, $L = 1150$ nm, and $P = 1400$ nm. Thicknesses of the silicon nitride membrane and gold film are 100 nm and 50 nm, respectively.

[43,44]. The dual-resonant behavior ($\lambda_1 \sim 1490$ nm and $\lambda_2 \sim 3800$ nm) shown in Fig. 3a is due to the superposition of the spectra of the constituting vertical (Fig. 3b) and horizontal (Fig. 3c) H-shaped apertures. In the composite aperture system, these individual responses shift to shorter wavelengths, where $\lambda_{vertical} = 1740$ nm and $\lambda_{horizontal} = 3900$ nm, respectively. Magnetic field intensity distributions ($|H|^2$) calculated at the dual-resonances in the composite system clearly demonstrate the origin of the dual-resonant behavior, i.e., at λ_1 (λ_2), highly enhanced local electromagnetic fields mainly concentrate at the top and bottom aperture arms (left and right aperture arms).

5. Quantification of nearfield intensity

Fig. 4a and b shows the electric field intensity distribution ($|E|^2$) for the dual-modes at λ_1 and λ_2 , respectively. For the mode at shorter wavelengths, highly enhanced local electromagnetic fields are mainly concentrated at the top and bottom aperture arms with an intensity enhancement factor as large as ~ 500 . In contrast, for the mode at longer wavelengths, they

localize along the long axis of the horizontal aperture, with an intensity enhancement factor of ~ 900 . **Fig. 4c** and d shows the yz -cross-sectional profile for the electric field intensity enhancement calculated at $x = 100$ nm (where the electric field intensities are maximum), demonstrating that highly enhanced local electromagnetic fields are homogeneously distributed along the aperture wall through z -direction, while their intensities gradually decreases along y -direction.

We then quantitatively demonstrate the accessibility of these local electromagnetic fields, which is highly important for applications demanding strong nearfield properties. Here, we integrate the electric field intensity within a volume (V), where the intensity drops to a cut-off value. Since the maximum intensity for the mode at λ_1 is smaller than the one at λ_2 , we set $1/e^2$ of the electric field intensity of the mode at λ_1 as the cut-off [45]. **Table 1** shows volume and the corresponding integral of the nearfield intensities calculated within it. Here, the mode at λ_2 supports ~ 18 times larger volume. Considering the additional effect of the larger nearfield intensity enhancement, the mode at λ_2 has ~ 20 times larger integral value compared to the mode at λ_1 .

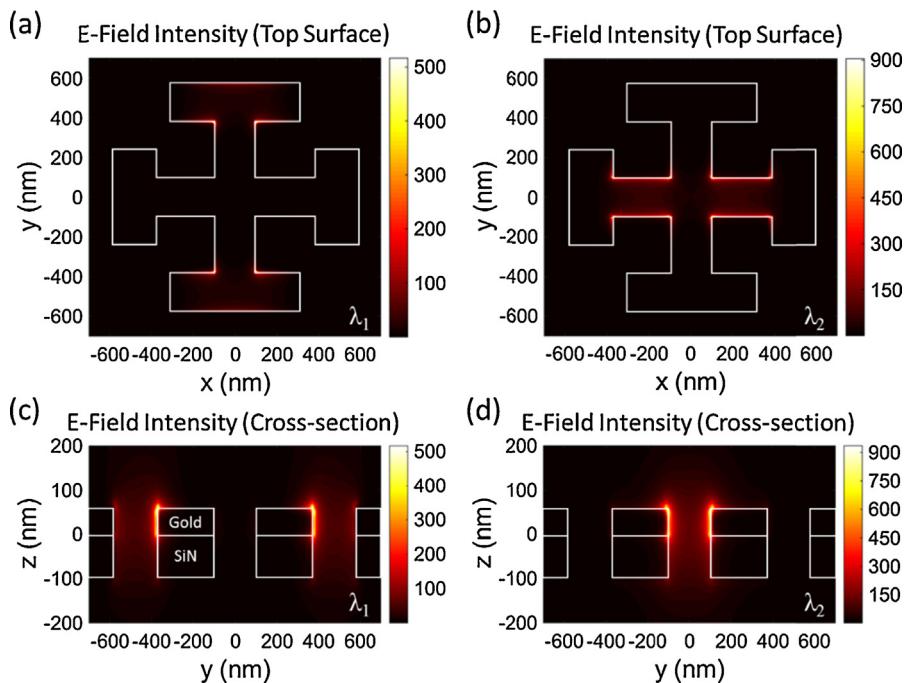


Fig. 4. Electric field intensity enhancement distribution ($|E|^2$) for the JC-shaped aperture calculated at $\lambda_1 = 1490$ nm and $\lambda_2 = 3800$ nm (a,b) at the top gold surface and (c,d) through cross-section. Corresponding device parameters are: $s = 600$ nm, $g = 470$ nm, $w = 200$ nm, $L = 1150$ nm, and $P = 1400$ nm. Thicknesses of the silicon nitride membrane and gold film are 100 nm and 50 nm, respectively.

Table 1
Cut-off volume and the integral of the electric field intensity enhancement over the cut-off volume for the mode locating at λ_1 and λ_2 .

	Cut-off volume (V)	$\iiint v \text{Intensity} \times dV$
Mode at λ_1	$64,500 \text{ nm}^3$	49,182
Mode at λ_2	$1,197,250 \text{ nm}^3$	989,500

6. Fine-tuning of dual-resonant spectra through the aperture geometry

Fig. 5 summarizes the dependence of the dual-resonant spectral response on geometrical parameters. Fig. 5a shows the variations within the plasmonic modes for different L values, where other parameters are kept constant at: $s = 600$ nm, $g = 470$ nm and $w = 200$ nm. Spectral response of the rectangular shaped apertures has a linear dependence on their side, which is perpendicular to the polarization direction of the illumination source as investigated earlier [46]. Therefore, for the mode at λ_2 where the local electric fields are concentrated at the horizontal rectangular aperture along the x -axis, changing L significantly alters this mode and shifts it to longer wavelengths for larger L . In contrast, for the mode at λ_1 , since there is no field localization at the vertical H-shaped aperture, we do not observe any spectral variations. Fig. 5b

shows the variations within the plasmonic modes for different g values where other parameters are kept constant at: $s = 600$ nm, $L = 1150$ nm and $w = 200$ nm. As for the mode at λ_2 , local magnetic fields are concentrated at the left and right aperture arms, increasing g shifts this modes to longer wavelengths. On the other hand, as the mode at λ_1 localizes at the aperture arms along the vertical direction, varying g does not alter this mode. Fig. 5c shows the variations within the plasmonic modes for different s values where other parameters are kept constant at: $g = 470$ nm, $L = 1150$ nm and $w = 200$ nm. Varying s has an opposite effect compared to g on spectral variations. Here, as for the mode at λ_1 , local magnetic fields are concentrated at the top and bottom aperture arms, increasing s shifts this modes to longer wavelengths. In contrast, as the mode at λ_2 localizes at the aperture arms along the horizontal direction, varying s does not alter this mode. Here, even though changing s and g results in identical variations in the aperture geometry, the spectral shifts within the mode at λ_2 (due to the variations in g) are larger than those within the mode at λ_1 (due to the variations in s). This is due to the larger intensity integral supported by the mode at λ_2 , which is directly proportional to the sensitivity of the system to the change in the surface conditions. Fig. 5d shows the variations within the plasmonic modes for different w values where other

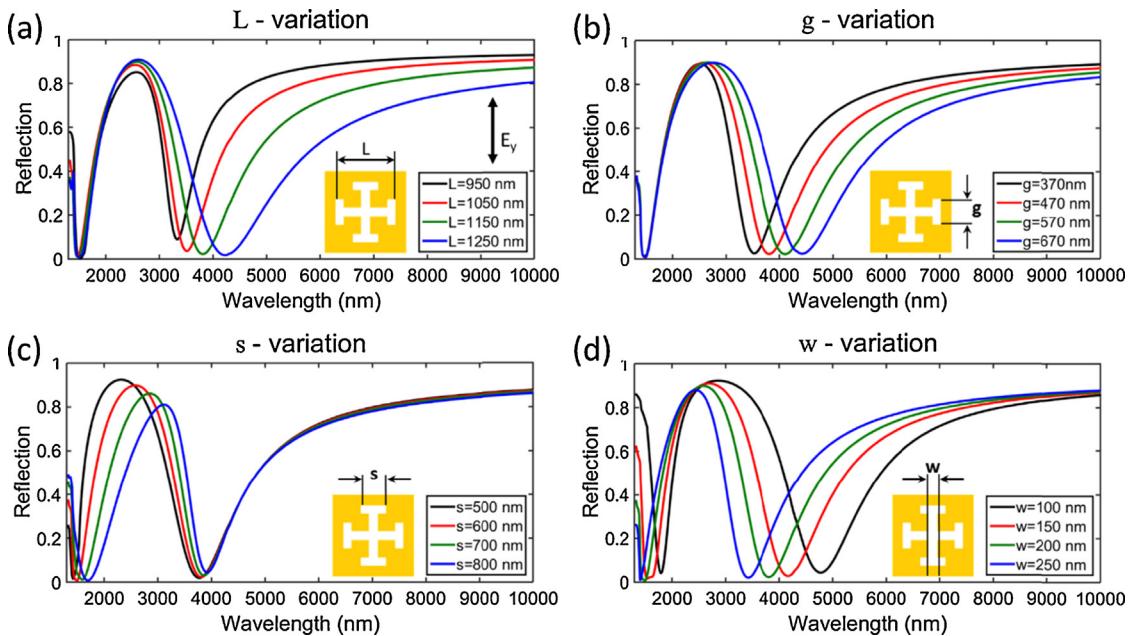


Fig. 5. Spectral variations within the calculated reflection spectra of the JC-shaped aperture for different geometrical parameters. (a) L -variation, where $s = 600$ nm, $g = 470$ nm, $w = 200$ nm. Figure inset shows the polarization direction of the illumination source. (b) g -variation, where $s = 600$ nm, $L = 1150$ nm, $w = 200$ nm. (c) s -variation, where $g = 470$ nm, $L = 1150$ nm, $w = 200$ nm. (d) w -variation, where $s = 600$ nm, $g = 470$ nm and $L = 1150$ nm. Thicknesses of the silicon nitride membrane and gold film are 100 nm and 50 nm, respectively, and $P = 1400$ nm.

parameters are kept constant at: $s = 600$ nm, $g = 470$ nm and $L = 1150$ nm. Here, both modes shift to shorter wavelengths for larger w values, where the spectral shift for the mode at λ_2 is larger than that of at λ_1 , which is due to two factors. First, increasing this parameter decreases the length of the horizontal aperture, at where the local electric fields are mainly concentrated for the mode at λ_2 . Second, even though changing w varies all aperture geometry identically, as the mode at λ_2 supports larger integral value, it varies stronger compared to the mode at λ_1 . Therefore, the results presented in this section clearly show that by changing the geometrical parameters, we can precisely tune the spectral responses for realizing aperture systems working in the desired wavelength ranges.

7. Sensitivity of the dual-resonant response

Supporting highly accessible large local electromagnetic fields, JC-shaped aperture system significantly improves light-matter interaction in nanometer scale, which is highly important for surface-enhanced applications demanding strong nearfields [47–49]. As an example to highlight this advantageous nearfield characteristic, we demonstrate numerical calculations on refractive index sensitivity, where the aperture system

is embedded in different refractive indices of bulk solutions: DI-water ($n = 1.3325$), acetone ($n = 1.3591$), isopropyl alcohol, IPA ($n = 1.3776$) and glycerol ($n = 1.4722$). Fig. 6a shows the variations within the spectral response of the aperture system, demonstrating that as the refractive index increases, plasmonic modes shift to longer wavelengths. Here, as the mode at λ_2 shows stronger sensitivities (due to larger near-field integral values) compared to the one at λ_1 , it shows larger spectral shifts. In order to show the sensitivity character of each mode, we calculate refractive index sensitivity, defined as the ratio between the change in the resonance wavelength and the change in the refractive index of the adjacent medium ($S = \Delta\lambda/\Delta n$). Fig. 6b shows the linear dependence of the resonance wavelength of each plasmonic mode to the refractive index of the bulk medium. These results clearly show that the mode at λ_2 (green line) shows stronger refractive index sensitivities (calculated by taking the derivative of each linear λ, n function) compared to the one at λ_1 (red line), ~ 2968 nm/RIU vs. ~ 1285 nm/RIU (RIU = refractive index unit). Supporting dual-resonances with line-widths (full-width-half-maximum, FWHM), 463 nm and 1161 nm for the modes at λ_1 and λ_2 , the aperture system support figure-of-merit values ($FOM = S/FWHM$), 2.8 and 2.6, respectively. These values are quite comparable

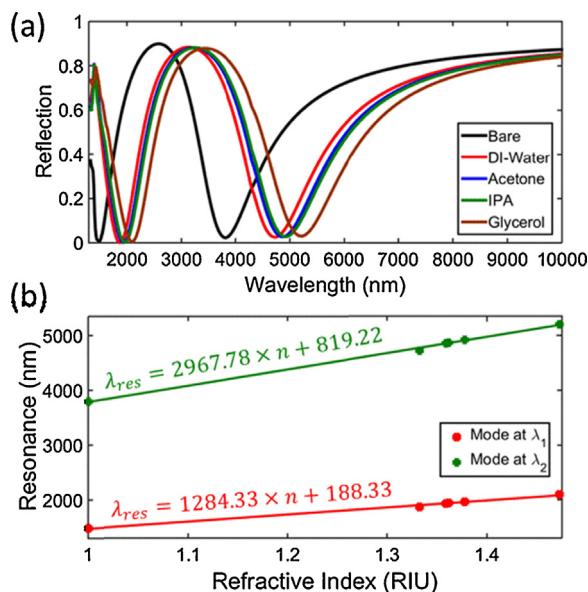


Fig. 6. (a) Spectral variations within the calculated aperture response for different refractive indices of bulk solutions: DI-water ($n = 1.3325$), acetone ($n = 1.3591$), IPA ($n = 1.3776$) and glycerol ($n = 1.4722$). (b) Linear dependence between resonance wavelengths (λ_{res}) of the dual-plasmonic modes and refractive indices. Corresponding device parameters are: $s = 600$ nm, $g = 470$ nm, $w = 200$ nm, $L = 1150$ nm, and $P = 1400$ nm. Thicknesses of the silicon nitride membrane and gold film are 100 nm and 50 nm, respectively.

with the state of the art label-free plasmonic biosensors designed for mid-IR region shown before [50,51].

8. Conclusion

In conclusion, we introduce a dual-resonant metamaterial based on Jerusalem cross (JC)-shaped nanoapertures. We numerically investigate the physical origin of the dual-resonant behavior which is due to the individual spectral response of the constituting aperture elements. For experimental realization, we utilize a high-precision and lift-off free fabrication method based on electron-beam lithography. We also introduce a fine-tuning mechanism for controlling the spectral response of the aperture system through geometrical device parameters. We perform refractive index sensitivity calculations in order to numerically show the aperture system's sensitive nature to the changes in the surface conditions. Finally, the quantitative analyses on the overlap between local electromagnetic fields and the medium in the vicinity of the aperture system show the good correlation between sensitivity of each mode and accessibility of optical fields. Possessing a multi-resonant spectral behavior with easily accessible large

local electromagnetic fields, JC-shaped nanoapertures can be a highly good candidate for large variety of surface enhanced optical applications.

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