Fano-Like Resonances in Split Concentric Nanoshell Dimers in Designing Negative-Index Metamaterials for Biological–Chemical Sensing and Spectroscopic Purposes

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In this study, we investigated numerically the plasmon response of a dimer configuration composed of a couple of split and concentric Au nanoshells in a complex orientation. We showed that an isolated composition of two concentric split nanoshells could be tailored to support strong plasmon resonant modes in the visible wavelengths. After determining the accurate geometric dimensions for the presented antisymmetric nanostructure, we designed a dimer array that shows complex behavior during exposure to different incident polarizations. We verified that the examined dimer was able to support destructive interference between dark and bright plasmon modes, which resulted in a pronounced Fano-like dip. Observation of a Fano minimum in such a simple molecular orientation of subwavelength particles opens new avenues for employing this structure in designing various practical plasmonic devices. Depositing the final dimer in a strong coupling condition on a semiconductor metasurface and measuring the effective refractive index at certain wavelengths, we demonstrate that each one of dimer units can be considered a meta-atom due to the high aspect ratio in the geometric parameters. Using this method, by extending the number of dimers periodically and illuminating the structure, we examined the isotropic, polarization-dependent, and transmission behavior of the metamaterial configuration. Using numerical methods and calculating the effective refractive indices, we computed and sketched corresponding figure of merit over the transmission window, where the maximum value obtained was 42.3 for Si and 54.6 for gallium phosphide (GaP) substrates.

Index Headings: Split concentric nanoshells; Plasmon hybridization; Fano-like resonance; Metamaterial; Figure of merit; FOM.

INTRODUCTION

Classical and quantum plasmonics have been considered fundamental concepts in characterizing various optic-based structures and devices in nanoscale dimensions for diverse applications, such as biological–chemical sensing, routing, superlensing, waveguiding, photovoltaic applications, and surface-enhanced Raman spectroscopy.1–5 In the past decade, artificial oligomer-type plasmonic molecules have received great attention due to their ability to support strong coupling of surface plasmon resonances in the nanoscale regime.6–8 The unique orientation, inherent symmetry, and strategic geometry of these nanoparticle-based, closely packed clusters allow for a tremendous intensification and hybridization of the excited electromagnetic (EM) fields inside the cluster system that can be described by plasmon hybridization theory.5,7 It is well understood that the hybridization of plasmon resonant modes includes supporting strong subradiant dark and superradiant bright resonant modes by mentioned subwavelength structures.7,8 Breaking the symmetry of a cluster causes inducing new dark modes at the energy continuum of the bright mode; therefore, the mathematical analysis of plasmon resonance hybridization becomes highly complex in antisymmetric nanostructures. Somewhere in between, the interference of opposite modes gives rise to Fano-like and Fano resonances along the scattering profile.9,10 The performance and quality of the Fano resonance dips are strongly dependent on the geometric, chemical, and environmental characteristics of a certain nanoparticle aggregate. For instance, when we illuminate a certain cluster using linear or azimuthal light sources, the behavior and quality of the Fano minimum that appears can be used in characterizing biological–chemical sensors with a certain sensitivity.

As another example, core–shell nanoparticles have extensively been employed in designing plasmonic structures and metamaterials due to the extraordinary tunability of the geometric parameters, tuning and shifting the plasmon resonances in the desired bandwidth, while the resonant modes can be intensified and localized robustly in the gap distance between the core and shell.15,16 In addition to sensing applications, the Fano resonance feature can be exploited in designing high-speed and accurate Fano switches or modulators, and optical wavelength (de)multiplexers.13,14

The shape and material of nanoparticles in a molecular nanocluster have significant impacts on the optical response and performance of the structure. When the optical response of various spherical nanoparticles were compared, researchers considered the nanoshell a strategic particle that provides outstanding geometric versatility and tunability due to large number of revisable geometric parameters.15,16 As another example, core–shell nanoparticles have extensively been employed in designing plasmonic structures and metamaterials due to the extraordinary tunability of the geometric parameters, tuning and shifting the plasmon resonances in the desired bandwidth, while the resonant modes can be intensified and localized robustly in the gap distance between the core and shell.15,16 In addition, researchers verified that two concentric nanoshells as a symmetric structure, called a nanomatryushka unit, yields an extra degree of freedom in its geometry and provides outstanding tunability in structural features compared to a single nanoshell or even core–shell nanoparticles. Furthermore, a symmetric nanomatryushka configuration can be tailored to support strong hybridized plasmon resonances from the visible to the near-infrared region.7

Recently, nanoparticle aggregates in periodic arrays that are deposited on metasurfaces have been extensively used to design negative refractive-index metamaterials due to their ability to support strong coupling of surface plasmon resonant modes in the visible wavelengths.5,6 The shape and material of nanoparticles in a molecular nanocluster have significant impacts on the optical response and performance of the structure. When the optical response of various spherical nanoparticles were compared, researchers considered the nanoshell a strategic particle that provides outstanding geometric versatility and tunability due to large number of revisable geometric parameters.15,16

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material structures for numerous purposes, such as sensing using high outstanding FOMs.7,19,20 Regarding the effect of metasurfaces on the transmission and absorption of optical power and excited EM fields, researchers have proposed and created numerous optimized devices, from sensing to superlensing, both theoretically and practically.

In this article, we study the plasmon response of a nanomatryushka unit composed of two concentric nano-shells in which the symmetry of the molecular structure is broken. Splitting both nanoshells with different split-gap angles, we determined and measured the appropriate geometric dimensions for the configuration being examined under excitation by transverse and longitudinal polarizations. Investigating the optical response of an isolated split concentric nanoshell, we show that the final artificial antisymmetric configuration presented interesting behavior when we adjusted two analogous units of this nanoparticle structure close to each other as a dimer. To illuminate the presented dimer in weak and strong coupling regimes, we calculated the spectral responses for the structure and depicted them using scattering cross-sectional profiles under different conditions. We verified that robust enhancements in the intensity and localization of the excited plasmonic fields can be performed and that, due to the symmetry missing in the structural properties and the appearance of dark plasmon modes, Fano-like dips can be observed along the scattering diagram. Depositing an array of the Au dimer on various metasurfaces in periodic arrays, we designed a metamaterial structure that can be employed in sensing applications with a high FOM.

THEORETICAL MODEL AND NUMERICAL METHOD

We employed the finite-difference time-domain (FDTD) method (Lumerical FDTD Solutions package) as a numerical tool to study and extract the optical properties of the split concentric nanoshells and to calculate the scattering efficiency profiles. To this end, we set the spatial cell sizes \((dx, dy, dz)\) to 2 nm, with the number of cells at 12 000, the simulation time at 6000 fs, and the number of snapshots at 17 690. Considering numerical stability for the employed components, we set the simulation time step to a \(dt\) of \(\sim 0.04\) fs, using the Courant stability condition. Moreover, perfectly matched layers (PMLs), with 32 compact layers, were used as the boundary conditions, with a high ratio of absorption of undesired scattered fields. Using these settings for the boundaries, we reduced the reflection of the destructive scattered fields to the workplace significantly. To excite surface plasmon resonances in the examined subwavelength structure, we used a linear electrical plane wave as an incident light source with the pulse length of 2.653 fs and an offset of 7.523 fs. In addition, we used the complex permittivity of Au nanoparticles with the experimentally determined Johnson–Christy constants,21 which can be explained by a monospecies Drude dispersion model:22

\[
\varepsilon' (\omega) + i \varepsilon'' (\omega) = \varepsilon_{Au} (\omega) = \varepsilon_{interband} - \frac{\omega_p^2}{\omega (\omega + i \Gamma)}; \quad \varepsilon_{interband} = \varepsilon_{\infty}
\]

where \(\varepsilon\) is the permittivity, \(\Gamma\) is the damping constant, \(\omega_p\) is the bulk plasma frequency, and \(\varepsilon_{interband}\) is the contribution due to the interband transitions. It should be noted that the value of \(\varepsilon_{interband}\) strongly depends on the operating wavelength and that \(\Gamma\) is determined by considering the structural sizes of the nanostructure. Thus, for an Au nanoshell with the radius \(R = 120\) nm and considering the calculation method for Drude parameters of Hövel et al.,23 we determined following values for the employed Drude theory: \(\varepsilon_{interband} = 9.72\), \(\omega_p = 9.244\) eV, and \(\Gamma = 0.0701\) eV. Moreover, we determined the frequency of onset for interband transitions \(\omega_{on\,interband}\) as 2.295 eV. Figures 1a and 1b exhibit the energy-level diagrams for the concentric nanoshells in symmetric and antisymmetric regimes that can be
used describe the plasmon hybridization theory between the split sphere and nanocavity. The major difference between the two diagrams is the charge density distribution in the nanoshells, which is entirely opposite in the two regimes due to the presence of split gaps. The other difference is correlated to the split sections in the nanoshells and nanosphere. In the antisymmetric regime, we removed the definite parts of the inner and outer shells with certain and different angles. Due to the geometric dependency of the optical response of an antisymmetric nanomatryushka structure, we were able to sketch the energy-level diagram of concentric nanoshells, including the interaction between a nanosphere and a nanocavity. Also, this geometric versatility increases the aspect ratio of the particles combination, and this feature can be considered to be meta-atoms when we design a metamaterial structure.

In addition, it is possible to express the dipole \( \mathbf{p} \) as an equation that contains electric polarizability \( \alpha \) and electric field \( \mathbf{E} \) as \( \mathbf{p} = \alpha \mathbf{E} \). Therefore, the total electric field can be considered to be \( \mathbf{E} = (1/\alpha)\mathbf{p} \), and the total electric field on the nanosphere and nanocavity can be written as:

\[
\left( \frac{1}{\alpha} \right) \mathbf{P}_{\text{sphere}} = \mathbf{E}_{\text{inc}} + \left( \frac{1}{\alpha} \right) \mathbf{P}_{\text{cavity}} \tag{2a}
\]

\[
\left( \frac{1}{\alpha} \right) \mathbf{P}_{\text{cavity}} = \mathbf{E}_{\text{inc}} + \left( \frac{1}{\alpha} \right) \mathbf{P}_{\text{sphere}} \tag{2b}
\]

To calculate the electric field on the nanosphere, we considered the Lorenz gauge to be time independent:

\[
A(x) = \frac{\mu_0}{4\pi} \int J(x') e^{ik|x-x'|/\varepsilon} d^3x' \quad \tag{3}
\]

where, \( k \) is the wavevector of the incident spectrum. By assuming \( |x-x'| = r \), where \( r \) is the intercenter distance between two nanoparticles, we can rewrite the vector potential in Eq. 3 as:

\[
A(x) = \frac{\mu_0}{4\pi} \frac{e^{ikr}}{r} \int J(x') d^3x' \tag{4}
\]

Consequently, the electric dipole field is:

\[
E = \frac{e^{ikr}}{4\pi\varepsilon_0} \left( 3n(n\cdot\mathbf{p}) - \mathbf{p} \right) \left( \frac{1}{r^3} - \frac{ik}{r^2} + k^2(n\cdot\mathbf{p}) \times \mathbf{n} \right) \tag{5}
\]

where \( n \) is the unit vector pointed from the center of the nanosphere to the center of the nanocavity.

As a result, for the near-field interaction between the sphere and the cavity, the electric field for each object can be simplified as:

\[
E = \frac{1}{r^3} \left[ 3n(n\cdot\mathbf{p}_{\text{sphere}}) - \mathbf{p}_{\text{sphere}} \right] \tag{6a}
\]

\[
E = \frac{1}{r^3} \left[ 3n(n\cdot\mathbf{p}_{\text{cavity}}) - \mathbf{p}_{\text{cavity}} \right] \tag{6b}
\]

Due to the polarization direction of the incident field \( \mathbf{E}_{\text{inc}} \), which is vertical to \( n \), Eqs. 6a and 6b can be simplified as:

\[
\mathbf{E}_{\text{sphere}} = -\frac{1}{r^3}(\mathbf{p}_{\text{sphere}}) \tag{7a}
\]

\[
\mathbf{E}_{\text{cavity}} = -\frac{1}{r^3}(\mathbf{p}_{\text{cavity}}) \tag{7b}
\]

This means the polarizability for the interaction, \( \alpha_{\text{int}} \), between the cavity and sphere can be considered to be \( -(1/r^3) \).

Also, the using proposed numerical method, we can calculate the polarizability of the cavity and sphere numerically. To this end, we employed the method verified by Bohren and Huffman, where the polarizability of a certain spherical nanoparticle can be obtained using Eq. 8:

\[
\alpha = 3V \varepsilon - 1 \frac{1}{\varepsilon + 2} \tag{8}
\]

where \( \varepsilon \) is the dielectric function, and \( V \) is the volume of the particle or object.

Finally, the plasmon hybridization between dipole fields of a sphere and cavity can be expressed using Eq. 9:

\[
\sum \mathbf{p}_{\text{cavity}} \times \mathbf{P}_{\text{sphere}} = \frac{1}{\varepsilon_{\text{sphere}} \varepsilon_{\text{cavity}}} - \frac{1}{\varepsilon_{\text{inc}}} \mathbf{E}_{\text{inc}} \tag{9}
\]

As a result:

\[
\sum \mathbf{p}_{\text{cavity}} \times \mathbf{P}_{\text{sphere}} = \frac{1}{\varepsilon_{\text{sphere}} \varepsilon_{\text{cavity}}} - \frac{1}{\varepsilon_{\text{inc}}} \mathbf{r}^3 \mathbf{E}_{\text{inc}} \tag{10}
\]

Inserting the volume of the nanocavity and the split sphere based on the geometric dimensions of the investigated split nanoshells, we used this method to calculate and sketch the scattering profile for the structure during excitation and hybridization of plasmon resonances. Note that, due to the modifications performed in the nanomatryushka structure and missing symmetry, the volume of the particle combination will be variable during the determination of the polarizability and scattering cross-sectional profiles.

**RESULTS AND DISCUSSION**

Figure 2a shows a two-dimensional schematic diagram of the examined nanomatryushka in symmetric and antisymmetric orientations; the geometric parameters of the split nanoparticles are indicated inside the diagram. The inner and outer radii for the interior and exterior shells are \( a, b, c, \) and \( d \), respectively; \( t \) is the space between concentric shells filled with a dielectric substance (e.g., silica); and \( \theta_1 \) and \( \theta_2 \) are the split angles of the inner and outer nanoshells, respectively. The influence of these parameters on the performance of the structure during exposure to incident light is extremely significant. As an important parameter, split angles change the polarizability of the examined
configuration (x) entirely, and this symmetry cancellation directly influences the dipolar momentums $d$ and $m$ of the electric field $E$ and magnetic field $H$, respectively. We have described the dependence of the polarizability on the volume of the particle as well as corresponding dielectric function (Eq. 8) in the previous section; hence, symmetry breaking gives rise to dramatic constructive or destructive effects on the excited EM fields, which must be controlled and applied correctly. For instance, for a complete and symmetric configuration of two concentric nanoshells based on our presented dimensions, the volume of the configuration is exactly

$$V = \pi h \left[ \left( b^2 - a^2 \right) \left( \theta_1 / 2 \right) + \left( d^2 - c^2 \right) \left( \theta_2 / 2 \right) \right].$$

Consequently, applied symmetry cancellation causes tremendous changes in the polarizability and, subsequently, in the scattered, refracted, and absorbed EM fields. Figure 2b shows the scattering efficiency profile for the concentric nanoshells in split regime with the default geometric dimensions under transverse and longitudinal polarizations. In this case, the split angles have significant effects on the scattering efficiency and the appearance of various dipolar and multipolar modes. Figure 2c illustrates two-dimensional snapshots of the plasmon resonance excitation inside the two concentric split nanoshells under both transverse and longitudinal polarizations. Examining these figures, we detect three noticeable peaks for dipolar and multipolar energies, at $\lambda = 670$ nm, $\lambda = 555$ nm, and $\lambda = 450$ nm. The difference in the energy and number of peaks between the two diagrams originates from the polarization direction of the incident light to the split sections in the nanoshells, and the superior behavior of the longitudinal polarization is obvious, especially when we consider the size of the red-shift of the generated peaks to the longer wavelengths and their energies. Figure 2b shows the direction of the incident light for different incidence polarizations. In this case, the split angles have significant effects on the scattering efficiency and the appearance of various dipolar and multipolar modes. Figure 2c illustrates two-dimensional snapshots of the plasmon resonance excitation inside the two concentric split nanoshells under both transverse and longitudinal polarizations that resulted from the numerical modeling simulations. Examining these figures, we detect a dramatic intensification in resonant modes energies, which corresponds to appearance of new dark modes as multipolar energy peaks. Also, these snapshots provide the number of modes under both opposite polarization excitations. One of the key effects that must be considered is the retardation or re-radiation effect. For this, we employed Mie scattering theory as a computational theory to calculate the scattering cross-sectional profiles. In this theoretical method, and also in the classic electrostatic model, changing the phase direction is not applicable for the multipolar modes and is considered only for the dipolar modes. Also, we verified...
that the retardation effect is highly significant in nanoparticle chains and complex structures, and that, for the proposed configuration, due to the simplicity of the structure, the effect of the phase direction is negligible.

To understand the effect of each geometric parameter on the scattering efficiency of the structure, we modified their sizes accurately and individually to probe the behavior of the configuration during these alterations and also to obtain accurate geometric sizes. Figure 3a exhibits the results of modifications in the geometric parameters of the inner nanoshell when the geometries of the surrounding shell were fixed. To this end, we first increased the size of \( a \) while \( b \) remained constant (\( b = 65 \text{ nm} \)) and the split angle was \( \theta_1 = 30^\circ \). Obviously, increasing the inner radius of the interior nanoshell directly decreases the thickness of the particle, and as a result, the positions of dipolar and multipolar extremes red-shifted to longer wavelengths for \( 30 \text{ nm} \leq a \leq 50 \text{ nm} \).

Considering a calculated profile for the structure, for \( a > 50 \text{ nm} \) the intensities of the dipolar and multipolar peaks are reduced dramatically, which originates from the incapacity of such a thin split nanoshell to support strong plasmon resonances. Next, we examined the effect of alterations in \( b \) while \( a \) remained constant (\( a = 40 \text{ nm} \)). Figure 3b shows the results of this modification on the spectral response of the structure. Increasing the size of \( b \) directly causes the thickness of the shell to increase to support the strong resonant modes efficiently. In the diagram, we can see a significant intensification in the energy of multipolar modes in addition to the dipolar peak energies for \( b = 75 \text{ nm} \) and \( b = 85 \text{ nm} \). This enhancement in the structure performance quality includes the appearance of high-order energy modes, initiated from the presence of the split sections in the structure. Technically, the incomplete circulation of resonant modes gives rise to robust interference between opposite modes of the inner and outer nanoshells, and the intensified plasmon resonance energy is localized in the gap distance of the split section. Controlling and directing this localized and reinforced energy would be helpful to induce strong plasmon resonances while employing the structure in the design of dimer and analogous molecular plasmonic configurations.

Worth noting here is that, when we increase the size of the outer radius to \( b > 85 \text{ nm} \), the space \( t \) between two split nanoshells decreases; this gives rise to dramatic decreases in the quality of the energy modes that appeared. Supporting the strong localization of surface plasmon resonances (LSPR) and producing interference between the excited electric and magnetic modes of two concentric nanoshells require a sufficient dielectric space \( (t) \) between them. Hence, when we scale the radii of the nanoshells down (or up), the gap size between two nanoshells must be kept at an appropriate dimension. From the results of our full numerical simulations of an optimal resonance condition, we set this parameter at \( t > 25 \text{ nm} \).

Next, we examined the behavior of the antisymmetric nanoparticles structure during modifications in the radii and size of the outer shell while the radii of the interior shell were fixed and the polarization of incident light was longitudinal. We modified the inner radius of the outer shell \( c \) in the range 120–140 nm. We detected similar behavior in the outer shell as in the interior nanoshell. Figure 3c presents a numerically calculated cross-sectional diagram of the scattering efficiency for the latest changes, where \( d = 160 \text{ nm} \). As we can see, when we incrementally increased the size of \( c \), the plasmon resonance frequency red-shifted to longer wavelengths, while the intensity of the dipolar and multipolar extremes increased dramatically. Obviously, these changes caused increases in the size of \( t \) and reductions in the thickness of the outer shell. When we compare the effects of the included parameters, the influence of
The thinner outer nanoshell is dominant due to its direct connection with the plasmon resonance oscillations, coupling, and hybridization; therefore, we observed a dramatic decrease in the quality of plasmon resonances for $c \geq 140$ nm. The effect of the outer radius of the exterior nanoshell, $d$, on the plasmon response of the split nanoshells is depicted in Fig. 3d, where the inner radius of the outer shell is constant ($c = 130$ nm).

Increasing the size of $d$ significantly red-shifts in the position of the dipolar and multipolar modes in the longer spectrum, and there is also an enhancement in the energy of the modes. This condition is valid for $160$ nm $\leq d \leq 200$ nm. In contrast, for $d > 200$ nm, due to the increases in the thickness of the exterior nanoshell, we observed a dramatic increase in the absorption ratio of the optical power, which gave rise to undesired loss (e.g., internal damping). As a result of these structural examinations, we red-shifted the peak of the dipolar and multipolar plasmon resonant modes to the visible spectrum efficiently. This feature will help us design plasmonic devices that are able to function effectively in this domain, such as optical routers and sensors.

The other important parameters that have major impacts on the plasmon response of the nanostructure examined are the split angles corresponding to the nanoshells ($\theta_1$ and $\theta_2$). Considering our numerically obtained simulation results, we altered these parameters individually to determine the appropriate angle for the split sections. Figure 4a exhibits the spectral responses of the proposed nanostructure during alterations in the angle of the split section of the interior nanoparticle ($\theta_1$), where the other geometric dimensions of the structure were set to the prior quantities and $\theta_2 = 45^\circ$. As a fundamental parameter, the split angle can be varied from $15^\circ$ to $120^\circ$; however, note that further decreasing the angle of the split gap caused the multipolar modes to disappear. In this regime, the effect of the antisymmetric features of the structure was reduced dramatically; therefore, in the earlier and further simulations, we set the split angle for both nanoshells to $\theta_2 = 30^\circ$. In contrast, increasing the size of the split gap caused the loss of some parts of the Au nanoshells, and the negative result is that the structure would not be able to support strong plasmon resonances due to the limited metallic area or surface for plasmon oscillations. Consequently, by satisfying an appropriate trade-off among the geometric parameters, we were able to optimize the intensity of robust plasmon resonances and also the energy of the dipolar and multipolar extremes. Examining the diagram in Fig. 4a, we find that the appropriate angle for the inner nanoshell is in the range $30^\circ \leq \theta_1 \leq 60^\circ$. As we expected, increasing the size of the split gap in the interior, or smaller, nanoshell intensifies the energy of plasmon modes and leads to a remarkable enhancement in the performance of the structure for the angle interval. This improvement includes a red-shift in the plasmon modes to the longer spectra, where the energy of the higher-order modes is increased and multipolar shoulders appear in the scattering profile. For $\theta_1 > 60^\circ$, a dramatic decrease in the energy of the plasmon resonance is reported, which includes a blue-shift to shorter wavelengths and disappearance of the high-order modes.

Here, due to the size and the unique role of the exterior nanoshell in supporting LSPR modes, modifications in the split-gap angle $\theta_2$ for this part of antisymmetric nanomatryushka has an intense effect on the spectral response of the structure. Figure 4b shows the results of these alterations when the other geometric parameters remain fixed. In the earlier study, to extract the plasmon response of the proposed structure, we employed an Au nanoshell with the split-gap angle of $\theta_2 = 30^\circ$. When we decreased the size of $\theta_2$, we observed a significant red-shift in the position of the dipolar peak while the energy of multipolar modes was reduced and even disappeared because of the produced symmetry and the energy of the dark modes decayed significantly. In contrast, when we increased the size of the split-gap angle, this provided a required antisymmetric structure; hence, the energy of subradiant dark modes increased. Despite of this symmetry cancellation, the position of superradiant and subradiant extremes red-shifted to longer wavelengths, which includes distinct shoulders of the multipolar modes along the scattering diagram.

Considering the profile sketched in Fig. 4b, we see that this enhancement in the performance of the structure is valid for $60^\circ \leq \theta_2 \leq 85^\circ$. For the split gap with the angle of $\theta_2 \leq 90^\circ$, a strong localization of plasmon modes is required due to the insufficient surface of the Au shell and, consequently, the hybridization of plasmon resonant modes cannot be implemented efficiently; hence, the energy of the modes were reduced in a way similar to the interior nanoshell. Finally, considering the results of all the earlier numerical trials, we determined the optimal structure to have the following geometric dimensions: $a = 40$ nm, $b = 80$ nm, $c = 130$ nm, $d = 185$ nm, $\theta_1 = 45^\circ$, $\theta_2 = 85^\circ$, and $t = 50$ nm.
Next, extending the number of split nanoshell units to design a dimer nanostructure, we deposited two nanoparticle units in close proximity to each other, with a gap of a few nanometers. Figure 5a shows two diagrams of the dimer composed of split nanoshell particles, where the split sections of both particles are positioned face to face. This orientation of the particles yields an outstanding opportunity to intensify and enhance the plasmon resonances at the split sections, which can be described by the plasmon hybridization theory. The gap space between two particle units is an edge to edge distance ($D_d$), which can be used for the strength of the plasmon resonance energy coupling. To provide a detailed study, we considered numerically the optical response of the proposed dimer in both weak and strong coupling regimes.

Using previously measured geometric sizes for the examined structure, we deposited particle units onto a glass substrate with permittivity $\varepsilon = 2.1$ and gap distance $D_d = 35$ nm (weak coupling). Illuminating the structure using a linear plane wave under transverse and longitudinal polarizations, we calculated numerically the scattering efficiency profile for the structure. Figure 5b shows the scattering cross-sectional profile for both polarizations for the weak-coupling regime. Examining the full numerically calculated scattering profile, we observed two distinct extremes associated with the dipolar and quadrupolar modes with significant energies along the diagram at wavelength $\lambda$ of $\sim600$ and $1050$ nm, respectively, for the transverse polarization mode. Also, we detected a shallow dip between two distinct shoulders in the range 675–800 nm that cannot be considered to be a product of dark and bright mode interference. Due to the low strength of the coupling between particles and also the direction of the incident light polarization (parallel to the split gaps), a complete and efficient hybridization of the plasmon resonance modes was not performed. Figure 5b also contains the spectral response of the structure during illumination using longitudinal polarization; here, we detected two distinct extremes at $\lambda$ of $\sim1000$ and $625$ nm, for the dipolar and quadrupolar peaks, respectively. In addition, there was a noticeable dip between the sub- and superradiant modes; as in the previous case, this cannot be considered a product of the destructive interference between dark and bright modes. Note that the energy of the opposite modes is higher than under transverse polarization due to the light source being perpendicular to the split gaps. Therefore, the possibility of the appearance of a Fano-like dip is high in a simple system of two split-shell units. Figure 5c contains two-dimensional snapshots for the investigated dimer under transverse (i) and longitudinal (ii) polarizations. Examining in these pictures, we can perceive a significant localization of plasmon resonances at the edge of the split sections, specifically for the longitudinal polarization mode.

In contrast, positioning two concentric nanoshell units closer to each other, reducing the edge to edge distance to $D_d = 12$ nm, we illuminated the structure to monitor its behavior in the strong coupling regime. All the modes appeared distinctively in the space between the two nanoparticles. Figure 6a exhibits the scattering cross-sectional diagram for the strong coupling condition under transverse and longitudinal polarizations, where two distinct extremes with significant energies have been generated for the longitudinal polarization (perpendicular to the split gap). An interesting feature here is the appearance of a Fano-like dip at $\lambda = 810$ nm, while the positions of sub- and superradiant modes red-shifted.

![Fig. 5](image-url)
to longer wavelengths. Therefore, by choosing a correct design of the dimer and performing a highly accurate analysis, a Fano-like minimum can be achieved in this simple dimer. Figures 6b(i) and 6b(ii) illustrate in two-dimensional snapshots the tremendous localization of excited surface plasmon resonances at the split sections of nanoshells, where the charge distribution direction in inner and outer nanoshells are in opposite azimuths, which gives rise to interference between excited dark and bright modes of the two nanoparticles; the result of this process is a Fano-like dip. Figure 6c illustrates the charge density distribution direction through the dimer structure under longitudinal polarization excitation. The noteworthy point here is the opposite direction of the charge distribution inside the inner and outer nanoshells of an individual nanoparticle configuration, which is correct for the formation of a Fano resonant mode. We can employ this ability of the proposed systematic dimer—composed of Au concentric split nanoshells in a strong regime—in designing metamaterial structures that have the potentials to be used in highly accurate plasmonic-sensing applications. The geometric versatil-
substrate as a metasurface with a set thickness can be used in numerous applications. 

Material structures with negative refractive indices that can provide low-loss, polarization-dependent, and isotropic metamaterials. Consequently, we can exploit this in designing meta-atoms with robust plasmon resonances in the visible domain; hence, we can consider each split-shell dimer as a meta-atom in designing metamaterial configurations. The negative electric and magnetic polarizabilities were obtained in the position of dipole moments for the final dimer structure (Figs. 7a and 7b, respectively). Using the novel theoretical method proposed by Asadchy et al.28 We computed the electric and magnetic polarizabilities for the final dimer structure (Figs. 7a and 7b, respectively) using the novel theoretical method proposed by Asadchy et al.28 The negative electric and magnetic polarizabilities were obtained in the position of dipole moments at a distance of 850–1100 nm. We extracted the optical properties as follows. Using the transmission window, we computed the effective refractive index \(n_{\text{eff}}\) at a position of 1000 nm. This condition provides us with an opportunity to consider the dimer structure as a meta-atom in designing metamaterial configurations; the strong polarization dependency of the dimer verifies this.

To design a metamaterial structure, we must calculate all the fundamental parameters, and the electric and magnetic polarizabilities are the most important factors in these configurations. Using our proposed theoretical method for studying the nanostructure, and placing the dimer on an n-type Si substrate (metasurface), we computed the electric and magnetic polarizabilities for the final dimer structure (Figs. 7a and 7b, respectively) using the novel theoretical method proposed by Asadchy et al.28 The negative electric and magnetic polarizabilities were obtained in the position of dipole moments around \(\lambda\) of \(\sim1000\) nm. This condition provides us with an opportunity to consider each split-shell dimer as a meta-atom with robust plasmon resonances in the visible domain; consequently, we can exploit this in designing low-loss, polarization-dependent, and isotropic metamaterial structures with negative refractive indices that can be used in numerous applications.

To this end, we deposited final dimer onto a Si substrate as a metasurface with a set thickness \(h_{a} = 1000\) nm and refractive index of \(n = 3.48\). Figure 8a contains a three-dimensional diagram of the metamaterial structure composed of certain arrays of subwavelength dimers, where all the dimers are placed at a set distance from each other to prevent the possible near-field coupling effect. This diagram also includes a detailed description of the geometric parameters; the propagation direction \(k\) is indicated by an arrow. The overall size of the proposed structure is \(4.5 \times 4.5\) \(\mu\)m. Illuminating the structure using a linear plane wave source and defining the transmission window for the metamaterial at a \(\lambda\) of \(\sim850–1100\) nm, we extracted the optical properties as follows. Using the transmission window, we computed the effective refractive index \(n_{\text{eff}}\) at a \(\lambda\) of \(\sim945\) nm to be approximately \((-1.315 + i0.0427)\), and using the corresponding FOM \(= |\text{Re}(n_{\text{eff}})/\text{Im}(n_{\text{eff}})|\), we determined the value of FOM to be \(-30.8\). Further investigations showed that the FOM strongly depends on the wavelength of the transmission window interval; for instance, for at a \(\lambda\) of \(\sim1025\) nm, FOM is \(-38.4\). Figure 8b presents a two-dimensional snapshot of the power absorbed by the nanoparticle dimers. 

Figure 8c indicates that the real part of the \(n_{\text{eff}}\) determined over the transmission window is negative. Figure 8d shows the numerically calculated FOM diagram over the transmission window for the final metamaterial structure. Examining in this diagram, we can determine the value of the computed FOMs for various wavelengths; the maximum possible FOM obtained is \(\sim42.3\) at a \(\lambda\) of \(\sim1090\) nm.

After evaluating recent and analogous structures using the proposed metamaterial configuration, we found the superior behavior of the structure studied here to be obvious because of the high value of FOM and its transmission window. More recently, various structures have been suggested based on core–shell nanoparticle arrays or split rings deposited on various semiconductors, such as Si, indium phosphide (InP), and gallium phosphide (GaP), which are lower in quality than the metamaterial structure proposed here.14,29,30 We evaluated the performance and sensitivity of the metamaterial structure based on dimer arrays composed of Au split nanoshells on various metasurfaces; Fig. 8e exhibits a comparative diagram of the calculated FOMs for metamaterials structures in which we changed only the substrate of the configuration. As we can clearly see, the FOM obtained using GaP, \(\sim54.6\), is bigger than the FOMs for the other metasurfaces.

We have shown that, due to the significant impact of the size of the split gap and its position on the performance of the structure, the direction of the incident light and its polarization are highly important. On the basis of the simulation results, using longitudinal polarization as the incidence mode resulted in a remarkable enhancement in the plasmon resonance intensification and hybridization, which resulted in a pronounced Fano-like resonance in the simple dimer structure. Therefore, we expected the same results for the metamaterial structure composed of split nanoshell dimer units. To verify this hypothesis, we depicted the transmission diagram for the two different polarizations with the polarization angle tolerance of \(\Delta \phi = \pm15^\circ\). Using this method, we were able to verify the depen-

Fig. 7. Polarizabilities over the wavelength variations, including double resonances. (a) Normalized electric polarizability. (b) Normalized magnetic polarizability.
dency of the negative-index metamaterial structure on the polarization direction, which is a crucial factor in defining the isotropy of the structure. Using the FDTD numerical method, we computed and sketched the transmission amplitude over the incident light angle at \( k \) of \( \sim 1090 \) nm for three different substrates (those corresponding to the highest FOMs) in Fig. 8f. Employing three semiconductor substrates with diverse refractive indices allowed us to examine comprehensively the effect of the substrate on the transmission amplitude of the proposed metamaterial structure. The result was that the Si and GaP substrates exhibited superior behavior. Note that the isotropy feature was performed for the metamaterial using the GaP substrate. For the other substrates and for longitudinal polarization, the transmission of optical energy and resonance coupling are clearly dramatic, while for the transverse polarization, the transmission amplitude is lower. Nevertheless, this condition does not affect the isotropy feature of the structure.

CONCLUSION

In this article, we investigated the plasmon response of a novel orientation of concentric nanoshells as an antisymmetric nanoscale configuration. We verified that the split nanoshells configuration we studied is able to support strong plasmon resonances in its nanosize dimensions. Using the scattering cross-sectional profiles, we determined the optimal geometric dimensions of the structure to obtain enhanced localization in the plasmon resonance energy. Adjusting the proximity of concentric split-nanoshell units to each other, we studied the hybridization of plasmon resonances and the appearance of dark and bright modes. We have shown that, in the strong coupling regime, the proposed dimer structure is able to support strong subradiant dark and superradiant bright modes, and that the weak interference between them results in the formation of a Fano-like dip in this simple molecular orientation. We then deposited the dimer onto a metasurface, and we calculated the transmission window and the effective refractive indices at desired wavelengths. Using this method, we determined numerically the corresponding FOMs and that the highest FOM (42.3) was achieved at a \( \lambda \) of \( \sim 1090 \) nm for a Si metasurface. Evaluating the effect of various metasurfaces on the transmission amplitude, we have shown that Si and GaP reflect the required isotropy and polarization dependency. When we compared the performances of these metasurfaces, we obtained the highest possible FOM for the metamaterial structure based on the GaP metasurface. The nanostructure studied here can be tailored for use in various applications, such as superlensing, switching, sensing, cloaking, nonlinear spectroscopy, and spintronics.

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