Fano Resonances in Complex Plasmonic Necklaces Composed of Gold Nanodisks Clusters for Enhanced LSPR Sensing

Arash Ahmadivand, Saeed Golmohammadi, Mustafa Karabiyik, Graduate Student Member, IEEE, and Nezih Pala

Abstract—Plasmonic nanoparticles in complex clusters and in specific molecular orientations are able to support strong plasmon and Fano resonances in their nanoscale geometries. In this paper, we examine the spectral response of a symmetric necklace shape nanostructure composed of Au nanodisk heptamers that are located in a close proximity to each other. Determining the appropriate geometrical parameters for the proposed necklace, we analyzed the effect of geometrical variations on the Fano resonance position and quality by calculating the scattering cross-sectional profile, numerically. Considering the strong localization of surface plasmon resonances (LSPR) in the heptamer clusters, the LSPR sensitivity for the studied necklace has been determined. Moreover, we evaluated the performance of the structure for different medium conditions by plotting corresponding figure of merit (FoM). To this end, we measured the plasmon resonance energy differences (∆E eV) over the refractive index (n) alterations, we quantified the corresponding FoM for the final necklace as 13.7, which can be utilized in designing precise and highly sensitive sensors. Ultimately, we proved that highly complex structures composed of metal nanoparticles clusters yield high sensitivity to the environmental perturbations.

Index Terms—Plasmonics, complex necklace, heptamer, Fano resonances, LSPR sensitivity, figure of merit (FoM).

I. INTRODUCTION

CONTROLLING the scattering of an incident optical energy during light-matter interaction and also, engineering the features of near-field localization of plasmonic nanostructures configurations have significant impacts on development of highly efficient subwavelength optical devices such as biosensors [1], [2], photovoltaics and solar cells [3], [4], nanoantennas [5], [6], surface-enhanced Raman spectroscopy (SERS) [7], and precise bio/chemical sensors [8]. Recently, self-assembled clusters in numerous simple and complex orientations (include symmetric and antisymmetric regimes) composed of several shapes of noble metallic nanoparticles have broadly been utilized in optic-based applications from sensing to switching [9], [10]. Gold, silver, copper, and aluminum in dimer [11], [12], trimer [13], quadrumer [14], heptamer [15], tetramer [16], octamer [17], and oligomers [18] designations are some of the important clusters that have wide-range of utilizations in designing various subwavelength all-optical devices. Each one of elucidated nanostructures reflects its own unique collective coherent free electron oscillations or plasmon resonance modes which are originated from the intense interaction of these modes between proximal nanoparticles in an individual cluster. To excite plasmon resonance energies inside a certain particle of a cluster, an intense incident optical energy is required, and these oscillations strongly depend on the corresponding phase of the plasmon oscillations inside the nanoparticles of a certain assembly. It is shown that surface plasmons (SPs) are classical oscillators and their resonances directly connected to the frequency of the incident optical energy [19], [20]. Additionally, closely spaced proximal nanoparticles have been considered as the structures that can be tailored to support strong electromagnetic plasmon resonances and Fano resonances that can be described by plasmon hybridization theory [21], [22]. The quality and intensity of these resonances strongly depend on the physical and chemical characteristics that are associated to the structural properties, substance of the particles and surrounding medium. It is well understood that symmetry breaking, geometrical alterations in the size of nanoparticles of clusters, changes in the permittivity of the host substance, and perturbations in the refractive index of the dielectric environment are some of the crucial factors that influence the intensity and quality of plasmon and Fano resonances dramatically [23]–[25]. The sensitivity of these nanoclusters to dimensional and environmental perturbations provide an opportunity to design accurate devices based on robustly coupled nanostructures. Conventionally, nanodisks with determined sizes and given substances have extensively been used to design compact and complex clusters with an identical gap sizes between proximal nanoparticles as $D_{nb}$, where $n$ is the number of the identical nanoparticles in an isolated heptamer cluster. The plasmon response of these subwavelength structures includes superradiant bright and subradiant dark resonance modes, and a weak coupling between these modes leads to appearing of a pronounced Fano dip in the scattering efficiency profile of the configuration.

The quality and behavior of Fano minima can be employed in designing localized surface plasmon resonance (LSPR)
TABLE I
FDTD PARAMETERS AND SETTINGS

<table>
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<th>Parameters</th>
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sensing systems and biodetection devices. Considering the geometrical features of a nanoparticle aggregate, spherical particles and cavities are the major type of nanostructures that have a wide range of utilization in designing elucidated nanoparticle clusters, and due to the coupling between excited dipolar and quadrupolar resonance modes regarding sub- and superradiant plasmon modes, pronounced Fano dips in single and multiple regimes can be formed. From the technical point of view, the quality of formation of Fano resonance in symmetric and antisymmetric structures can be different. Breaking the symmetry of a nanosize structure causes interference and coupling between strong superradiant dipolar modes and subradiant multipolar modes, the result of this coupling is a minimum at the spectral response of the structure. It is well accepted that symmetry missing gives rise to appearing of new dark modes along the scattering profile and controlling these modes are highly difficult. Therefore, there are a strong desire to yield Fano dips in symmetric clusters without encountering with symmetry breaking expenses. To realize this claim, numerous shape of nanoparticles with specific substances have been used. Nanodisks, rods, shells, matryushka, and pyramids are some of the popular nanoparticle clusters that can be tailored to support strong plasmon and Fano resonances in cluster type aggregates [11], [14]. Each one of mentioned particles provides specific geometrical features and tunability. Herein, we use the nanodisk particles in designing a necklace structure.

In this study, we investigate the optical properties and spectral response of a super cluster contains of six heptamer clusters composed of Au nanodisks with experimentally determined Johnson-Christy constants [26]. Proposed super structure is shaped in a necklace shape nanostructure (resembles oligomer-like orientation without central particle or particles unit). Also, we examine the plasmon response of the proposed nanostructure during structural modifications to investigate the Fano resonance behavior. In this method, we determined the appropriate geometrical sizes for the necklace based on Fano resonance behavior. It is also shown that Fano resonance in necklace shape configuration exhibit dramatic shifts to changes in the refractive index of the environmental dielectric substance. The LSPR sensitivity of the cluster aggregate is examined by plotting corresponding linear figure of merit (FoM). To this end, three-dimensional Finite-Difference Time-Domain (FDTD) method (Lumerical FDTD Solutions 8.9) is employed as a numerical tool to extract the unique properties of the proposed structure accurately [27]. The simulation parameters and settings are listed and described in Table. 1. This table also, contains the properties of the dipole light source.

II. RESULTS AND DISCUSSION

Figure 1(a-i) exhibits a schematic diagram of a simple symmetric heptamer composed of seven identical Au nanodisks with the radii of $R$. Figure 1(a-ii) shows nanodisk heptamers that are oriented in a necklace shape structure, and accordingly, six peripheral symmetric clusters in heptamer orientation are located in a circular fashion and deposited on an infinite glass substrate with a permittivity of $\varepsilon \sim 2.1$. Considering the structural features of the symmetric necklace structure, two neighbor heptamer nanoclusters have two adjacent nanodisks which supply the intense resonance coupling between proximal nanodisks. The polarization direction and the place of source is indicated by an arrow.

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On the other hand, Prodan et al. [21] proved that Fano resonances in symmetric clusters can be analyzed...
and characterized by plasmon hybridization theory, and Lassiter et al. [25] demonstrated that in the efficient near-field coupling regime, there are two modes regarding Fano resonance interferences which are connected to the bonding bright and antibonding dark plasmon resonance modes. Studies have shown that one of the Fano interferences is the coupling of light with superradiant bright mode (bonding plasmon mode), and as a result, the oscillation direction of the dipolar plasmon resonances in all of the cluster particles are same (In-phase mode), in contrast, in a subradiant dark mode regime (antibonding plasmon mode) which is related to the dipolar moment of the middle particle, the oscillation direction of the central and peripheral particles are in an opposite direction (Out-of-phase mode). Technically, in the quasistatic, nonretarded limit, there is no any net dipole moment in the subradiant dark mode and therefore, this resonance cannot coupled to light efficiently. On the other hand, in the retarded limit, the dark mode remains subradiant and the bright mode changes to superradiant mode and consequently, an interaction between dark and bright modes based on near-field plasmon resonance coupling can be performed. This robust interaction between sub- and superradiant plasmon modes induces a Fano resonance in the superradiant continuum at the superradiant energy mode [21], [25]. Here, we depicted the scattering cross-sectional diagram for the configuration during modifications in the structural geometries of utilized nanodisks during illuminating by an incident electric dipole source that is located in the center of the circular necklace. The noteworthy point here is the noticeable symmetry of the nanostructure, which leads to reflection of an isotropic optical response and Fano resonance. It should be noted that in the absence of central nanoparticle unit, the incident optical power is transporting in circular direction along the heptamers and subsequently, the circular orientation of the necklace helps to excitation of SPs and Fano-like resonances in each one of heptamers and coupling to the nearest one. The result of this strong coupling is a pronounced Fano minimum. Figure. 1(b) exhibits the scattering efficiency for the geometrical variations in the size of nanodisks of a necklace which is scaled up from the nonretarded limit to the completely retarded limit. It is obvious that, using nanodisks with the identical radii of \( R = 50 \) nm and the gap distance of \( D = 12 \) nm, a dipolar plasmon resonance is appeared at \( \lambda \sim 960 \) nm and a minima corresponds to the Fano resonance is occurred at \( \lambda \sim 1110 \) nm. Increasing the size of nanodisks radiuses in the range of 50-100nm, a dramatic red-shift has been observed in the Fano resonance position to the longer wavelengths (NIR spectra). This movement of Fano resonance includes an enhancement in the quality of Fano minimum, while it becomes deeper and narrower, and the reason comes from the ability of particle to support strong plasmon resonances. However, it should be noted that the negative side of this size increment to enhance the Fano minimum is the undesired absorption of incident electromagnetic fields, hence, for a necklace composed of Au nanodisk heptamers for the size of \( R > 100 \) nm a dramatic reduction in the quality of Fano resonance is expectable. Therefore, finding an accurate and correct deal between necklace geometries will help us to design a structure that is extremely useful for efficient optical purposes from precise sensing to fast switching. For instance, for nanodisks with the radii of \( R = 75 \) nm, 85nm, and 100nm, a Fano minimum has been induced at \( \lambda \sim 1300 \) nm, 1430nm, and 1560nm, respectively. Inset snapshots (Fig. 1(b) (i-iii)) show two-dimensional (xy) snapshots of plasmon resonance excitation and coupling between proximal heptamers and enclosed Au nanodisks. The most important point for the largest necklace (\( R = 100 \) nm) is that the subradiant dark mode emits slightly which causes to a trivial broadening of the Fano minima as well as unfavorable absorption of light by big particles. It is clear that controlled increments in the size of nanodisks of the necklace structure causes robust hybridization of plasmon resonances between adjacent particles, and as a result, this intense hybridization of high order plasmon resonant modes suppress the superradiant mode and in contrast, the subradiant mode red-shifts strongly. Moreover, the superradiant bright mode red-shift weaker than dark mode as can be seen inside the scattering cross-sectional figure. The origin of this phenomenon related to the mode orders of each one of the energy levels, for instance, the superradiant bonding mode contains dipolar modes while, the subradiant antibonding modes includes dipolar, quadrupolar, and even higher order of plasmon modes (multipolar plasmon resonance modes).

In continue, we examine the effect of gap distance variations between proximal nanoparticles of nanodisks clusters on the optical response of the nanonecklace. In prior studies, this parameter was set to a default value (\( D = 12 \) nm), and Figs. 2(a) and 2(b) exhibit the influence of increases and decreases of gap distance parameter on the Fano resonance position and quality, respectively. Accordingly, increasing the size of gap distance, the Fano minimum directly blue-shifts to the shorter wavelengths while the Fano resonance depth reduces and becomes broader. For instance, for the \( D = 16 \) nm and 17nm, Fano resonances has been induced at \( \lambda \sim 850 \) nm and 725nm, respectively (see Fig. 2(a)). This blue-shift of Fano resonance resulting by the occurrence of weak coupling regime in distant particles orientations. This regime refers to the interaction between adjacent particles results in slight splitting of the nanodisk modes merely into bonding (bright mode) and antibonding (dark mode) energy levels, and even more increment in the gap distance causes to disappearing of Fano dip and higher order of plasmon modes and eventually, only dipolar plasmonic resonance remains as a major extreme. In contrast, decrements in the gap distance strongly red-shifts the Fano dip to the larger spectra and resulting a desired deep and narrow dip (see Fig. 2(b)). For instance, for \( D = 6 \) nm and 5nm which refer to the strong coupling regimes, the Fano minimum induced at \( \lambda \sim 1850 \) nm and 1920nm, respectively. In this condition, each one of closely spaced nanoparticles induces a mixture of different plasmon bonding and antibonding modes with different energy levels. This hybridization of modes includes dipolar and multipolar moments and it is obvious in the diagrams that the multipolar resonances red-shift much more strongly than dipolar modes and the reason is the surpassing of dipolar moment by higher order interactions. Noticing in Fig. 2(b), changing the coupling regime from weak to robust condition, multipolar peaks become appear along the
Fig. 2. Scattering cross-sectional diagram is illustrated for the necklace of nanodisk heptamers while the gap distance between adjacent particles is altering. (a) increases in the gap distance resembles weak coupling regime which blue-shifted the Fano minima to shorter spectra which cause to induction of shallow and narrow Fano minima. (b) decreases in the gap distance resemble the strong coupling regime which red-shifted the Fano minima to larger spectra and the minima become deeper and broaden and (c) charge density direction through the heptamers for superradiant bright mode (i) and subradiant dark mode (ii) are demonstrated by arrows. Dual head arrows indicated the coupling between closely spaced heptamers.

scattering spectral profile. Figure. 2(c) illustrates the charge density distribution direction through the nanonecklace to for superradiant bright and subradiant dark modes by arrows. For instance, for a necklace with recent geometrical sizes and 5nm of gap distance, the superradiant extreme has been induced at $\lambda \sim 1765$nm and the subradiant extreme has been appeared at $\lambda \sim 2005$nm. Accordingly, in the bright plasmon resonance mode, the direction of charge distribution is in the same direction in all of the nanodisks of clusters (see Fig. 2(c)-i), and on the contrary, in the dark mode the charge density flowing is different through the disks (see Fig. 2(c)-ii). The charge density distribution inside the nanoparticles of clusters and between neighbors particles of clusters are indicated by arrows inside the diagram, therefore, proximal nanoparticles regarding two heptamer clusters play fundamental role in supplying required Fano interference by inducing and coupling Fano-like interference. From the physical point of view, each one of the clusters of the super-necklace acts like an individual simple nanoparticle, but the major difference here is the direction of the charge distribution, and in view of that the dipolar moments directions inside the heptamers are different in each one of clusters. Due to the presence of heptamers and structural complexity, the bright and dark modes can be supported efficiently, and the occurred interactions are highly severe due to the strong hybridization of resonance modes which supports deep and narrow Fano resonances. Considering the inherent and outstanding symmetricity of the necklace, the key point here is the intense interactions between adjacent clusters that causes appearing deep Fano minimum without symmetry breaking expenses and difficulties. The potential application of this necklace is using in array as a metamaterial structure for enhanced THz sensing and analogous approaches.

As we mentioned, proposed symmetric necklace provides the ability to obtain isotropic Fano dip. To verify this claim, we examined the spectral response of the complex necklace during variations in the incident polarization direction from $\varphi = 0^\circ$ to $90^\circ$. Figure. 3(a) demonstrates the scattering cross-sectional profile for polarization variations, while the necklace geometries have been set to optimized values as following: $R = 90$nm, $h = 80$nm and $D = 5$nm. As we expected, a deep and high quality Fano resonance is appeared at $\lambda \sim 1950$nm for all of three polarizations with a subtle red and blue-shift that does not affect the performance of the necklace noticeably. Figures. 3(b) and 3(c) exhibit two-dimensional snapshots for the nanonecklace under excitation with two different polarization directions ($\varphi = 0^\circ$ to $90^\circ$). Obviously, due to the symmetric nature of the necklace structure, fundamental alterations in polarization direction
Fig. 3. (a) Calculated scattering cross-sectional diagram for proposed nanonecklace composed of Au nanodisk heptamers under illumination by transverse to longitudinal polarization modes ($\phi = 0^\circ, 45^\circ, 90^\circ$). A pronounced Fano dip has been appeared at $\lambda \sim 1950$ nm with minor deviations in the position for different polarizations, (b) and (c) two-dimensional snapshots of plasmon resonance excitations in nanonecklace under polarization angles with $\phi = 90^\circ$ and $45^\circ$, respectively.

of the incident light did not affect the performance of the structure.

Next, we study the LSPR sensitivity of the examined necklace composed of Au nanodisk heptamers. This application of Fano resonance has broadly been investigated and applied in theoretical and experimental approaches of designing precise and high sensitive plasmonic sensors [28]–[30]. Earlier works have shown that simple clusters of nanoparticles are sensitive to the modifications in refractive index of the dielectric environment and appearing of narrow and high quality Fano resonance allows for exact determination of minor variations in the dielectric medium by shifting the resonance peaks [31], [32]. In order to evaluate the LSPR efficiency and sensitivity of a plasmonic subwavelength and finite structure, corresponding figure of merit (FoM) must be plotted and quantified. FoM is defined as a proportion of the plasmon resonance energy ($\Delta E$ eV) deviations over the dielectric environment refractive index ($n$) variations, which is divided by the width peak of the spectral response [25], [31]. Moreover, it is shown that for the Fano resonances in antisymmetric regime, the resonance energy is the midst point between the energy of the initial maximum and the last minimum, in addition we are able to define the width as the resonance energy margin between mentioned spectral extremes. Therefore, we evaluate the LSPR sensitivity of the necklace configuration of heptamers by employing FDTD simulation model which is used to plot the linear FoM figure. To this end, we applied the dielectric environment substances as following subsequently: Methanol (CH$_3$OH) $n = 1.331$, Ethanol (C$_2$H$_5$OH) $n = 1.361$, Butanol (C$_4$H$_{10}$O) $n = 1.399$, and Acrylic matching liquid $n = 1.491$ (based on presented data in Cargille Laboratories) [33]. During calculations, we assumed that the necklace of heptamers is completely immersed in the utilized dielectric liquids thoroughly, and the geometrical sizes of two examined structures are considered as nanodisks with the radiuses of $R = 90$nm (small structure) and 100nm (big structure), and the gap distance is $D = 5$nm for both of the structures. Figures. 4(a) and 4(b) illustrate the scattering cross-sectional diagram for two structures, respectively, while the dielectric medium is varied. For both of the structures the Fano resonance is red-shifted to the larger spectra by increasing the refractive index of the dielectric substance of surrounded ambient. For the air medium, the Fano resonances are appeared at $\lambda \sim 1520$nm and 1560nm, for smaller and bigger structures, respectively, while slight differences between Fano dips are easily evident. Noticing in both of the figures, Fano resonances for the presence of $n = 1.491$ are detected at $\lambda \sim 1790$nm and 1850nm for smaller and bigger structures, respectively. The corresponding red-shift of Fano resonance for the necklace with the disk radius of 100nm is much more than smaller one, but the Fano minimum for smaller necklace is deeper and narrower than the bigger one, which leads to sensing with more accuracy. The noteworthy point here is that this condition for smaller refractive indices is confirmed. As a result, the smaller necklace provides precise LSPR sensitivity for various dielectric environments. To provide comprehensive study, we plotted and compared the FoM figure for both of the examined optimal necklaces to quantify the LSPR sensitivity for both of the nanostructures. Obtaining the slope of the linear line for the Fano resonance energy over the refractive index alteration (0.72 and 0.67 for small and big necklace, respectively) and dividing by the Fano width (0.053eV and 0.051eV), the FoMs are determined as 13.7 and 13.1 for small and big necklace, respectively (see Figs. 4(c) and 4(d)). These results prove that the LSPR sensitivity and accuracy of the smaller necklace with the disk radii of 90nm is more than
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Fig. 4. LSPR sensitivity for two complex necklaces in small and big regimes are presented, (a) LSPR sensing for the smaller structure based on nanodisks heptamers with the radii size of 90 nm is investigated numerically based on FDTD simulation method. The Fano minima is red-shifted and get narrower by increasing the refractive index of the dielectric environmental substance, (b) LSPR sensing for the smaller structure based on nanodisks heptamers with the radii size of 100 nm is examined. The Fano minima is red-shifted to larger wavelength by increasing the refractive index of environmental substance, but the Fano is not as narrow as smaller necklace, and (c) and (d) Linear plots of the LSPR shifts of the Fano resonance over the refractive index of environmental substances to determine the FoMs for small and big nanonecklaces.

III. Conclusions
In this study, we investigated the optical properties and spectral response of a complex and highly symmetric arrangement composed of six nanodisk heptamers that are oriented in a necklace shape. We showed that the proposed plasmonic configuration provides a pronounced Fano resonance along the scattering efficiency profile during illuminating with a dipole source with transverse to longitudinal polarizations. The effect of structural modifications on the spectral response of the proposed necklace has been examined numerically. It is shown that increasing the size of nanodisks radii gives rise to red-shift of the Fano dip to the longer spectra (NIR). Increasing and decreasing the size of gap distance between proximal nanoparticles of complex arrangements causes blue and red-shifting of Fano resonances and resembles the weak and strong coupling regimes, respectively. Ultimately, we determined and quantified the LSPR sensitivity of the structure while the refractive index of the dielectric environment is a variant parameter. Therefore, plotting the linear figure for energy differences of Fano dips over the refractive index alterations for both assumed smaller and bigger necklaces, we determined the FoM for each structure as 13.7 and 13.1, respectively. Comparing proposed complex necklaces with similar simple structures, we realized that due to appearing of pronounced and high quality Fano minimum, the LSPR sensitivity of the examined heptamer-based structure is remarkable and superior which can be employed in designing precise Fano-based bio/chemical and gas sensors, and SERS subwavelength devise.

REFERENCES


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