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Influence of plasmon destructive interferences on optical properties of gold planar quadrumers

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Abstract

Arrays of planar symmetric gold quadrumers consisting of a central nano-disc surrounded by three similar nano-discs belonging to the D_{3h} point group were designed and fabricated. Since the geometrical configuration of quadrumers is the same as planar trigonal molecules, nano-discs can play the roles of artificial atoms to study the coupling trends among them. The plasmonic properties of the nano-disc structures are investigated by reflection spectrum measurement and finite-difference time-domain calculation with good agreement. Plasmon interaction among the nano-discs is also studied via a mass–spring coupled oscillator model. A pronounced Fano resonance (FR) is observed for the fabricated nano-discs with inter-disk gaps of around 18 nm during light irradiation at normal incidence. Although the obtained FR is independent of the excitation polarization, the near-field energy spatial distribution can be flexibly tuned by the polarization direction. This has potential applications in nano-lithography, optical switching and nonlinear spectroscopy.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

A study of artificial molecules with different combinations and arrangements has been experimentally undertaken recently [1, 2]. The coupling mechanism of each individual atom with the nearby atoms making up the molecules is the key to predicting the molecular characteristics [3]. It can contribute to the better understanding of molecule theories. Meanwhile, recent advancements in nano-fabrication provide the opportunity to design and fabricate nano-structures with arbitrary sizes and configurations. Out of several coupling mechanisms among the components of the nano-structures, plasmon coupling is a topic of intense current interest [4]. Localized surface plasmons are a result of the collective oscillation of the conductive electrons in the metallic nanoparticles [5]. Subsequently, an electromagnetic field gets excited and distributes some distance away from the particles. This is seen clearly in complex

nano-structures in which the plasmons interact with neighboring structures [6]. It has recently become apparent that the plasmons of metallic nano-structures, while describable by the classical electromagnetic theory, exhibit certain characteristics that are analogous to electrons in quantum systems [2, 7]. While bottom-up methods for artificial molecule creation, such as DNA scaffolding, have been investigated recently [8], a few researches have been aimed at studying plasmonic interactions in planar symmetric lithographically patterned artificial molecules, without a central disk or particle, like ring-like trimers, quadrumers, pentamers and hexamers [3, 9–13]. Among them, ring-like hexamers have been well studied due to the similar symmetry to benzene for unraveling the energy levels in conjugated hydrocarbon molecules [3]. But recent studies on symmetric planar structures with a central disk or particle, such as pentamers [14] and heptamers [3, 10–12], show that these structures can exhibit more interesting

behaviors, such as transition from the isolated to collective modes [3] and Fano resonance (FR) [10, 11]. Through group theory, it has been found that because of the symmetry in such oligomers, the optical responses are independent of the orientation of the in-plane polarization of light incident on the structure [12]. This capability encourages researchers to predict the characteristics of corresponding molecules with the same configurations. But the difference between planar structures with a central disk or particle and conjugated atoms in multi-gonal molecules is that the atoms in these molecules are typically non-co-planar. This is due to the tendency of surrounding atoms to achieve the maximum distance with respect to each other under a 3D configuration in order to provide the lowest energy for molecules to be in the stable state. CH_4 and PCl_5 are good examples of pentagonal and hexagonal molecules, respectively, in which the central atom and the surrounding atoms do not become placed in the same plane. Trigonal planar molecules are the only molecules with central and surrounding atoms which are co-planar and where all atoms are in the same plane symmetrically. But there is a lack of a comprehensive experimental study on this configuration.

In this work, arrays of planar symmetric quadrumers consisting of a central nano-disc and three outer ring-like nano-discs, which have D_{3h} spatial symmetry, are designed and fabricated. The recent study on plasmon modes in three nano-cylinder clusters was also related to irreducible representations of the point group D_{3h} [15]. The difference between the mentioned paper and our work is the existence of the central cylinder or disk. The plasmon interaction nature, among the central nano-disc and the surrounding ones in our configuration, promises to predict and allow us to understand the electronic states in trigonal planar molecules which have similar configurations, such as BF_3 , SO_3 and BCl_3 . Simultaneously, it can provide the qualitative basis to discover the energy levels of the molecular orbital in conjugated carbon atoms in a graphene plane or a carbon nano-tube. A detailed study on the plasmonic quadrumer is presented in which the near-field interaction among neighboring elements determines its optical response. More importantly, a pronounced FR is observed experimentally and predicted theoretically. This resulting FR is attributed to the destructive interference among anti-parallel dipole modes which can be observed independently of the excitation polarization at normal incident light. This is an improvement compared to previous studies, in which the FR was obtained by breaking the symmetry [16, 17], angling the incident light [18] or changing the excitation polarization [19]. This ability allows for a wider range of controlled optical applications, like surface-enhanced Raman spectroscopies, nano-antennas and biochemical sensors [20]. Meanwhile, it is shown that the resulting FR does not exist in the optical responses of individual nano-discs but appears when three identical nano-discs are placed around a central nano-disc belonging to the D_{3h} point group. This can be an analogue between the hybridization of the plasmons arising from each individual nano-disc in quadrumers and the same form of molecular orbital interactions from individual atomic orbitals in molecules [3]. Consequently, the study on collective modes

in the plasmonic quadrumer can provide more information on the complex electronic states in trigonal planar molecules. One more unique feature of the designed quadrumer is the localized near-field energy distribution in sub-20 nm gaps among the nano-discs. It is shown that the near-field energy distribution can be flexibly tuned by changing the excitation polarization, while the collective optical responses, such as the FR, are polarization-independent. This energy localization can be established for a normal incident light of a single source rather than co-illumination by two light sources with different incident angles or phase shifts [21, 22]. It can overcome the spatial restrictions of conventional optics with unique potential applications in nano-lithography [23] and nonlinear spectroscopy [22].

2. Experimental details

The arrays of monomers and quadrumers were fabricated by electron beam lithography (Elonix 100KV EBL system) on silicon substrates. A Ti film of 3 nm thickness was firstly deposited by an e-beam evaporator (EB03 BOC Edwards) on the substrate to increase the adhesion between the Au and the substrate. Then a 60 nm Au thin film was evaporated on the top of the Ti layer. Next, a negative electro-resist, hydrogen silsesquioxane (HSQ) (thickness: 50 nm), was spin coated. The samples were baked at $\sim 200^\circ\text{C}$ for 120 s. After the electron beam exposure and development, the nano-structures on the electro-resist were transferred down to the Au films by ion milling. To characterize the fabricated samples, a UV-vis-NIR micro-spectrophotometer (CRAIC QDI 2010 based on a Leica DMR microscope) was used. A normal incident light with linear polarization was applied to excite the nano-structures. Simulation was carried out by a three-dimensional finite-difference time-domain technique (FDTD) using a perfectly matched layer (PML) around the structures in the wavelength range of 300–1300 nm. The dielectric functions used for the simulation were obtained from the experimental data of Johnson and Christy [24].

3. Results and discussion

Figure 1(a) illustrates the molecular geometries of a H atom and planar trigonal molecules, and subsequently a gold monomer and a gold planar quadrumer formed as their plasmonic analogues, respectively. The formation of artificial molecules draws on the compelling analogy to the formation of molecules in chemistry. Figures 1(b) and (c) show the scanning electron microscope (SEM) images of the periodic patterns of the monomers and quadrumers fabricated on the silicon substrates and figure 1(d) plots the atomic force microscope (AFM) image of the quadrumer. The sizes and array configurations of the monomers were the same as those in the quadrumer array for the comparison and further analyses. The diameter and height of the nano-discs were 142 ± 1 nm and 60 ± 2 nm, respectively, and the gap between the nano-discs in the quadrumers was 18 ± 2 nm. Since the arising plasmons are extremely sensitive to the shapes of the nano-structures,

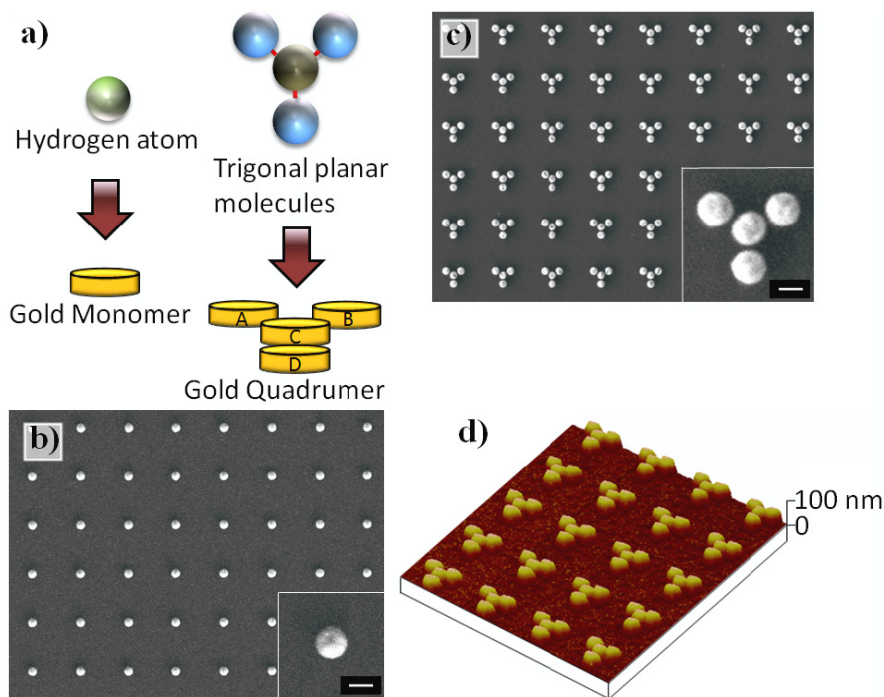


Figure 1. (a) Upper row: illustrations of the molecular geometries of a H atom and the trigonal planar molecule configuration; bottom row: their plasmonic analogues, a gold monomer and a gold quadrumer. SEM images of periodic array patterns of (b) monomers and (c) quadrumers. The scale bar is 100 nm. (d) AFM image of quadrumers.

significant efforts have been made to fabricate circular nano-discs in our experiments.

In order to investigate the hybridization among the nano-discs, the optical reflection responses of the plasmonic monomers and quadrumers were simulated as shown in figure 2(a). The measured reflection spectra are displayed in figure 2(b). It is apparent that the spectral profiles in figure 2(b) display the same characteristics as those simulated in figure 2(a) except for a 100 nm red shift of the experimental peak positions with respect to the calculated spectra. This is due to the absence of the substrates in the simulation. The reflection spectra of the Au monomers reveal the excitation of the dipolar mode which contributes to the emergence of a single resonance. But an interesting feature in the spectrum appears when three nano-discs are symmetrically placed in close proximity to the monomer, which functions as the central nano-disc. In this case, the mixed plasmons of the nano-discs hybridize and increase the chance to form anti-bonding and bonding plasmonic mode [25] transition. This is in direct analogy to the electron wavefunctions of atoms involved in trigonal planar molecules, which belong to the D_{3h} point group. The interaction of light and this gold quadrumer leads to the excitation of collective plasmons in the nano-discs. The net anti-parallel dipolar plasmon, arising from individual discs, hybridizes destructively, allowing the formation of a dark sub-radiant collective mode in addition to the bright super-radiant collective mode [26]. Because of this destructive interference, the peak position of the super-radiant mode cannot be exactly determined from the reflection spectrum. Consequently an asymmetric line-shape profile known as the FR [27], which was predicted in figure 2(a) at around 640 nm, can be seen

in the experimental measurement results in figure 2(b) around 750 nm as well. The existence of the FR in the quadrumers demonstrates that the condition of destructive interference between the sub-radiant mode and the super-radiant mode is sufficiently fulfilled. It is worth mentioning that, although the obtained FR shown in figures 2(a) and (b) is under x-polarized incident light, the reflection spectra for the other polarization directions are identical, as is expected from the symmetry considerations [12]. This occurs because the destructive interference in such structures is a result of interference among the plasmons arising from each individual component as well as gaps among them, whose scales are not comparable with the incoming light wavelength. That is why the quadrumer can be considered as an in-plane isotropic structure, in which the FR is achieved under totally symmetric conditions at normal incidence and also independent of the excitation polarization.

In order to elucidate the physics behind the appearance of the sub-radiant dark mode and the super-radiant bright mode, charge distributions at the respective simulated spectral positions of 640 nm and 780 nm are shown in figures 2(c) and (d), respectively. These plots are calculated for x-polarized incident light, keeping in mind that, due to the planar symmetry of quadrumers, the excitation polarization does not affect the optical response and it only leads to the rotation of the charge distribution pattern in all nano-discs simultaneously. The charge distribution pattern in a single quadrumer at a wavelength of 640 nm plotted in figure 2(c) shows the configuration of the anti-parallel dipole, which is responsible for the emergence of the sub-radiant mode. Although the disks A and B exhibit a rotated charge distribution pattern with respect to the central nano-disc C, the total oscillation

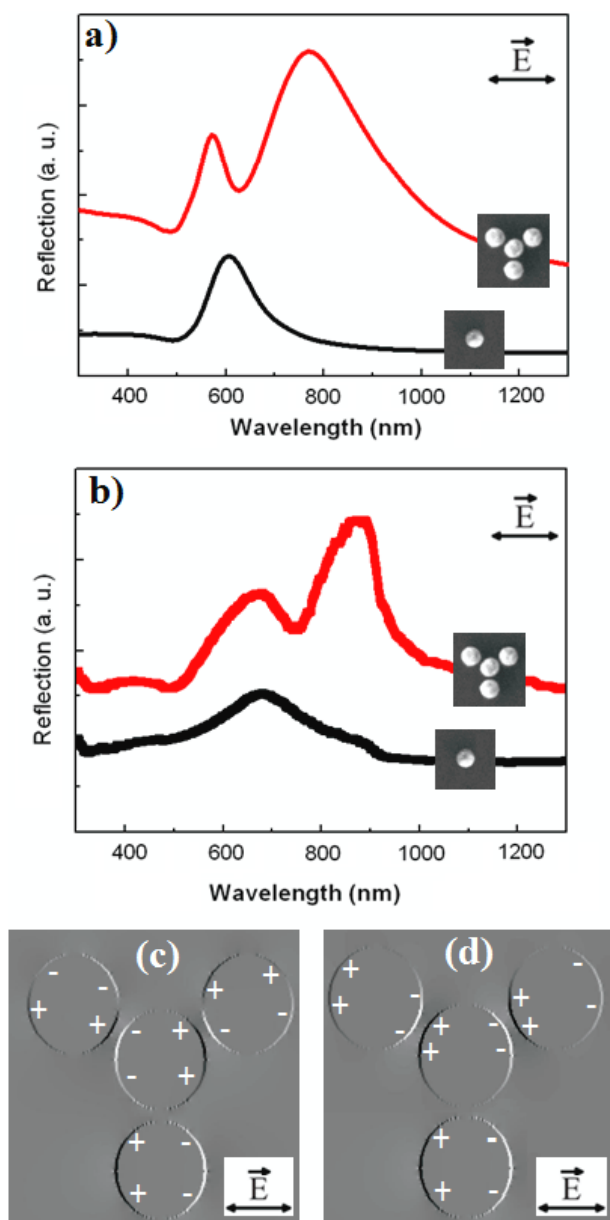


Figure 2. (a) Simulated and (b) measured reflection spectra of the monomers and quadrumers at x-polarized normal incidence. Calculated charge distribution of the quadramer at wavelengths of (c) 640 nm and (d) 780 nm by FDTD simulation.

directions of these two nano-discs are parallel to the oscillation direction of the nano-disc D. In other words, at this wavelength the dipole moments of the nano-discs A and B only boost the dipole moment of the nano-disc D, while the central nano-disc C oscillates totally out of phase. This central disk exhibits an anti-parallel dipole with respect to the total dipole moment of the other three nano-discs. Unlike this mode, the super-radiant bright mode in the quadrumers exists when the plasmons of all the four nano-discs oscillate in the same phase. The charge distribution pattern at a wavelength of 780 nm plotted in figure 2(d) represents the formation of the super-radiant mode. In this case, all the nano-discs within the quadramer structure experience strong coupling and form a collective

plasmonic mode. This is the wavelength at which the effect of the destructive interference decays totally. This destructive interference is also observed experimentally in the reflection measurement in figure 2(b) where the FR comes into existence at around 750 nm. This plasmon hybridization in quadrumers can be used to express the transformation among atoms with D_{3h} symmetry. It could be applied in an analogous manner to the molecular orbital theory [2] of trigonal planar molecules.

Furthermore, it is shown that although the obtained optical responses are independent of the excitation polarization, the spatial distribution of the near-field energy can be flexibly tuned by the polarization direction. Figure 3 shows the ability of the quadrumers to concentrate light down to sub-20 nm gaps. The localization of incident light has been investigated by means of co-illumination with two separate light sources, with different incident angles or phase shifts [21, 22]. But our study shows that the symmetry of quadrumers can lead to the energy localization of a single source with normal incidence. Interestingly, this spatial near-field localization can be tuned by the polarization, while the far-field spectral responses are polarization-independent at the same time. Since the characteristics of the metallic nano-structures exhibit certain characteristics that are analogous to electrons in quantum systems [2, 7], this ability of the quadrumers can overcome the spatial restrictions of conventional optics on atomic scales.

Figure 3(a) shows that the near-field energy localization can achieve several hundred-fold energy intensity enhancement within the gaps among the central nano-disc and the surrounding nano-discs of the quadrumers at a wavelength of 780 nm. While this localization can be obtained at a non-polarized incidence, the application of linear polarization gives more interesting localization patterns. Figure 3(b) plots the spatial energy distribution pattern with 30° excitation polarization with respect to the x -axis. It leads to the maximum near-field interaction within the gap between the nano-discs C and B. Consequently, a higher amount of energy is stored in this gap with respect to the two other gaps of the quadramer. Since the amounts of exhibited near-field energy within the two gaps of the nano-disc C to the nano-discs A and D are equal, this energy distribution can find a wide range of potential applications in plasmonics nanolithography [23]. These nanoscale triangular points of the localized energy with different intensities under one pulse of light irradiation can overcome the difficulties of sharp corner exposure [28]. Meanwhile, the energy localization pattern plotted in figure 3(c) reveals another potential application of the quadramer structure. This plot shows how the quadramer can be used as a planar optical switch [29] along three directions simultaneously. As can be seen, when a 60° excitation polarization is applied, the near-field energy is localized at two gaps out of the three gaps inside the structure. The third gap exhibits almost no near-field energy. This unique ability to localize the near-field energy in selected gaps between the nano-discs via excitation polarization provides the possibility to realize synchronized control systems in molecular nanoscale circuits [30].

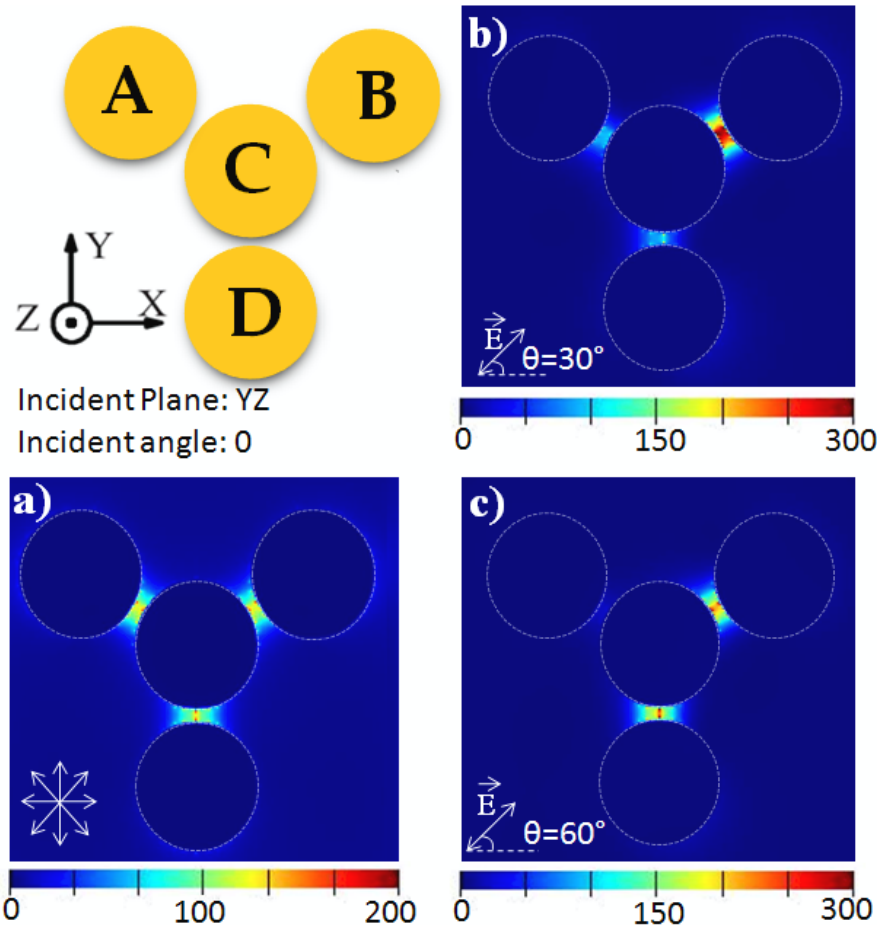


Figure 3. Calculated field distribution within the quadramer at x -polarized normal incidence at a wavelength of 780 nm: (a) non-polarized, (b) 30° polarization and (c) 60° polarization with respect to the x -axis.

4. Mass–spring mechanical model

The optical response of the quadramer is modeled with mechanical coupled oscillators. This can assist in the understanding the analogy between the plasmons in nano-structures and atomic and molecular wavefunctions, which is attributed to the fabrication of various shapes and sizes of the complex plasmonic nano-structures, which belong to the atomic and molecular configurations. The quadramer plasmonic system can be modeled by a three coupled interacting oscillators system, which is shown in figure 4(a). This physical model is an extension of the classical two oscillators system which was used to study the nature of the FR [31]. Our model is the simplified form from the 3D system consisting of four interacting oscillators to the 1D oscillator model, which is shown in figure 4(a). This simplification is based on two assumptions which can be obtained from the field distribution plots in figure 3. In these plots, it can be observed that the plasmons of the surrounding nano-discs do not interact with each other, unlike, their individual interaction with the central nano-disc. Meanwhile, in all polarization excitations, two out of the three surrounding nano-discs have a totally similar interaction with the central nano-disc. The oscillator 2 in the model shown in figure 4(a) represents these two disks. Meanwhile, the interaction of the third nano-disc

with the central nano-disc is different. Then the oscillator 3 represents the third surrounding nano-disc. Eventually the oscillator 1 is assumed as the representative of the central disk.

In the planar symmetric structures including the central nano-disc or particle, a dark mode exists when a nano-disc or particle is added to the center of the surrounding ring-like nano-discs or particles [3, 11]. This means that in the optical system, in the absence of a central nano-disc or particle, only the bright mode can be excited. Subsequently, in our analogous mechanical system only the oscillators |2⟩ and |3⟩ are driven by a periodic harmonic force $\mathbf{F}(t) = \mathbf{F}e^{-i\omega t}$ due to their responsibility for the appearance of the bright mode. This is analogous to the optical excitation of the bright super-radiant mode.

The motion equations of oscillators |1⟩, |2⟩ and |3⟩ are solved in terms of the displacements x_1 , x_2 and x_3 from their respective equilibrium positions in a one-dimensional system:

$$\ddot{x}_1(t) + \gamma_1 \dot{x}_1(t) + \omega_1^2 x_1(t) - \Omega^2 x_2(t) - \Omega^2 x_3(t) = 0, \quad (1)$$

$$\ddot{x}_2(t) + \gamma_2 \dot{x}_2(t) + \omega_2^2 x_2(t) - \Omega^2 x_1(t) = F e^{-i\omega t}, \quad (2)$$

$$\ddot{x}_3(t) + \gamma_3 \dot{x}_3(t) + \omega_3^2 x_3(t) - \Omega^2 x_1(t) = F e^{-i\omega t}, \quad (3)$$

where Ω is the coherent coupling frequency between interconnected oscillators, which represents the coupling

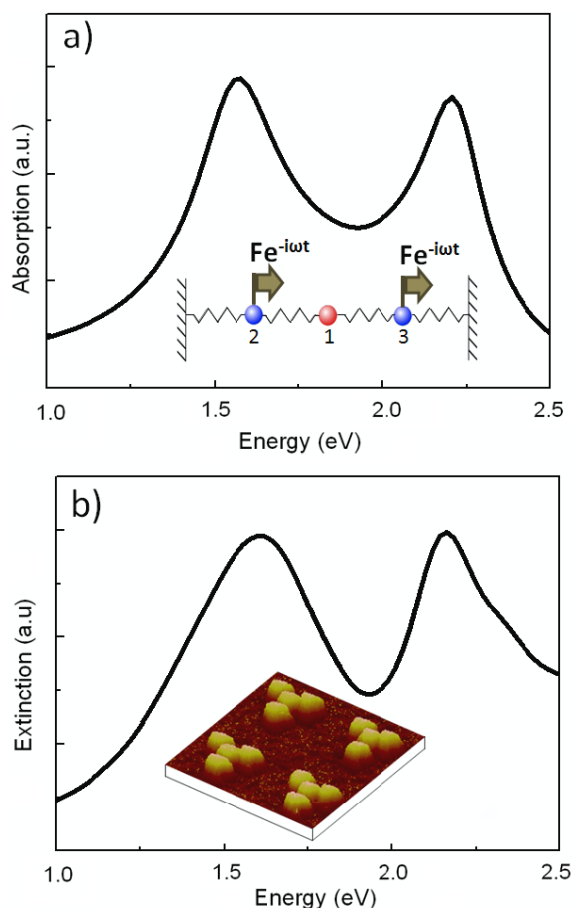


Figure 4. (a) Three coupled interacting oscillators representing the quadrumer optical responses and corresponding calculated power absorption, and (b) simulated extinction spectra of the quadrumer at x -polarized normal incidence by FDTD.

between anti-parallel modes in the optical system. γ_1 , γ_2 and γ_3 are the friction coefficients which are used to account for the energy dissipation of $|1\rangle$, $|2\rangle$ and $|3\rangle$, respectively. These friction coefficients represent the dissipated energy among the nano-discs during energy transformation at optical excitation. The mass values are assumed as $m_1 = m_2 = m_3 = 1$ and all the springs are the same. Meanwhile, the quadrumer bright plasmon mode is modeled with $\omega_2 = 1.9$ eV and $\omega_3 = 2$ eV as the representatives of the resonance frequencies of the three surrounding disks and the resonance frequency $\omega_1 = 1.9$ eV which represents the central nano-disc frequency. By tuning the coupling between modes ($\Omega^2 = 0.9$) and other parameters ($\gamma_1 = 2 \times 10^{-1}$, $\gamma_2 = 4 \times 10^{-1}$, $\gamma_3 = 5 \times 10^{-2}$ and $F = 1.5$), the absorption power of the oscillator system is calculated and sketched in figure 4(a). This is analogous to the optical excitation which reproduces the plasmon response of the quadrumer. Figure 4(b) shows the extinction spectra by FDTD simulation at x -polarized normal incidence. The absorbed powers of the oscillator system calculated by equations (1)–(3) and the FDTD simulated extinction are in good agreement. The dip position of both the absorption profiles in the mechanical model of figure 4(a) and the extinction profile in the simulation result in figure 4(b) is around 1.9 eV. This energy corresponds

to the wavelength of 652 nm, which is around the investigated maximum destructive interference for both the experimental and the simulated reflection spectra, as shown in figures 2(a) and (b).

5. Conclusions

The optical responses of the designed and fabricated plasmonic quadrumer were studied theoretically and experimentally. It is shown that the interference of plasmons arising from individual nano-discs of the quadrumer leads to several unique effects. The experimental results, the FDTD simulations and the mass–spring model are in good agreement. It is shown that the interference between the sub-radiant and super-radiant modes leads to a pronounced FR at normal incident light and independent of the excitation polarization. Furthermore, it is also demonstrated that the configuration of the quadrumer allows localization of the near-field energy selectively by changing the excitation polarization. This opens up potential applications in optical switching, nonlinear spectroscopy and nano-lithography. Meanwhile, the obtained results show that the distinct spectral features are possible in an ensemble of symmetric high-quality planar nano-lithographic structures with nanoscale gaps. The fabricated quadrumer provides an opportunity to draw inspiration for the design of artificial trigonal molecules using the plasmonic quadrumer, which can lead to the discovery of numerous useful electromagnetic analogues of chemical molecules in plasmonics. Particularly it can provide the opportunity for a better understanding of the molecular orbital theory of the planar trigonal molecules, such as BCl_3 , BF_3 and most importantly the carbon atoms within the graphene plane, where plasmons of neighboring structures or surfaces interact with each other. Therefore, the plasmons mix and hybridize just like the electron wavefunctions in similar atomic and molecular orbitals.

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