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Revealing the effect of plasmon transmutation on charge transfer plasmons in substrate-mediated metallodielectric aluminum clusters

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ABSTRACT

Aluminum nanoparticle nanocomplexes have extensively been utilized for sustaining ultrastrong plasmonic bonding and antibonding resonant modes across the ultraviolet to visible spectrum. In this study, we analyze the plasmon response for two conventional symmetric heptamer and antisymmetric octamer antennas mediated by conductive film as a substrate to induce very sharp Fano-resonant mode at the high energy states. Besides, presence of an underlying conductive film in touching regime with the plasmonic nanoantennas leads to formation of charge transfer plasmons (CTPs) across the deep-UV band. It is also shown that presence of dielectric carbon nanospheres in the gap spots between proximal nanodisks gives rise to breaking the symmetry of the assemblies, while the new magnetic multipolar modes are induced and divided the Fano dip in two parts as well as formation of a couple of charge transfer plasmon resonant shoulders. The compactness and geometries of the clusters allow for inducing substantially strong resonant modes across the deep-UV domain. Our investigations provide new pathways and features for designing multifunctional molecular probes, biochemical sensors, and cathodoluminescence antennas across the UV spectrum. The proposed analysis were done using a blend of Finite-Difference Time-Domain (FDTD) calculations and transfer of plasmonic charges in nanoscale systems.

1. Introduction

There have been extensive researches in the concept of plasmonic nanoparticle assemblies in both symmetric and antisymmetric orientations to support significant magnetic subradiant dark and electric superradiant bright modes as well as antisymmetric Fano resonances (FRs) via capacitive coupling of plasmons across the ultraviolet (UV) to mid-infrared region (MIR) [1–5]. These clusters have broadly been utilized for various purposes from biochemical sensing [6,7] to waveguiding [8,9]. It is also shown that metallic subwavelength antennas consisting of efficiently coupled nanoparticles can be tailored to provide remarkable absorption cross-section across a wide range of spectrum to be employed for several applications such as photodetection to photothermal spectroscopy [10,11], including substantial electromagnetic (EM) field enhancements in the offset gap spots between proximal particles. In this limit, the big portion of the absorption takes place at the position of spectral lineshapes or antisymmetric FRs, where the scattering extreme is suppressed and absorption feature enhanced, characterizing by plasmon hybridization theory [12–14]. Very recently, innovative methods have been proposed to intensify the energy of the photoexcited modes and also induce new plasmon resonant modes. It is strongly verified that placing metallic nanostructures in a quantum size gaps (smaller than 1 nm) apart, allows for supporting new plasmonic modes, called charge transfer plasmons (CTPs), happening at the energy states below the strongly hybridized resonances ($E < 0.9$ eV) that can be characterized by quantum tunneling effect, called Fowler–Nordheim tunneling [15,16]. The other method to induce CTPs is using direct transfer of charges between closely-spaced particles by connecting them with a conductive touching link such as nanowire [17]. In the limit, charges are able to shuttle between connected particles and provide the ability to support tunable and energetic CTPs and bright dipolar modes across the near-infrared region (NIR) to visible band. The latest technique to induce CTPs is depositing a conductive substrate below the metallic antenna in touching regime. In this case, transfer of charges between conductive substrate-mediated nanoparticles in an individual cluster

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gives rise to redistribution of plasmonic charges between nanoparticles and metallic film and leads to arising of CTPs [18]. As a recent achievement, the presence of a conductive aluminum layer under a plasmonic nanoparticle gives rise to narrowing the linewidth of the electric bright mode and enhancing the scattering efficiency at high energy states [19]. Using this facility, for plasmonic nanocomplexes, multipolar magnetic dark and dipolar bright modes can be blue-shifted to the higher energies, and the result of this suppression is inducing a narrow dip in between as a pronounced FR minimum. However, the effect of thin aluminum film on the induced CTPs by an integrated aluminum-based cluster across the UV spectrum is remained unclear. In recent years, capacitive coupling in aluminum nanoclusters in both symmetric and antisymmetric geometries has been employed for plasmonic biochemical detection [20,21] to colorimetric sensing applications [22]. Sustaining strong plasmonic FRs via hybridization of plasmons, aluminum was introduced as a potential substitution for expensive and rare metals for plasmonic and opto-electronic devices fabrication due to CMOS-compatibility, natural abundance, and absence of interband transitions across the UV band. It should be underlined that conventional noble metals (gold, silver, and copper) suffer from rapid oxidation and dissipative interband transition losses across the UV to the visible spectra. In contrast, despite of having a few nanometer of natural oxide layer around aluminum nanostructures, comparing the plasmonic response of aluminum structures with classical plasmonic metals at the mentioned domain confirms its superior plasmonic behavior [23,24]. However, one should not that fabrication of tightly compact aluminum subwavelength structures based on Mie-scattering theory [25] to operate at the deep-UV ($E > 4$ eV) needs for high resolution electron-beam photolithography machines or extreme-UV lithography techniques [26,27].

On the other hand, new techniques have been proposed to intensify the energy of dark modes for narrowing the lineshape of the plasmonic and coil-type FR modes along the visible to NIR by adding dielectric nanoparticles in the gap spot between proximal plasmonic particles in a certain cluster [28–30]. This technique also facilitates the fabrication difficulties in producing closely-packed nanoantennas without reducing the gap area between nanoparticles, which is providing highly intense plasmon modes analogous with subnanometer gaps. In the capacitive coupling regime, the result of this structural modification is inducing additional collective magnetic modes, or converting existing plasmonic modes to new ones, which lead to intensify the energy and quality of antisymmetric FR dip. In this limit, the spectral response shows a remarkable red-shift in the position to the longer wavelengths, while for the conductive junction a dramatic blue-shift was recorded to the shorter spectra [30].

In this study, we examine the plasmonic response for both symmetric and antisymmetric aluminum nanoparticle assemblies deposited on a conductive thin aluminum substrate to investigate formation of substrate-mediated CTPs and FRs at high energies. To this end, we employed symmetric seven-member heptamer and antisymmetric eight-member octamer nanoantennas that are strongly coupled to the underlying aluminum substrate. Inducing conventional plasmonic modes during illumination, we also monitored CTP resonances across high-energies. Breaking the symmetry of the clusters by insertion of dielectric carbon nanospheres in the gap spots between nanoparticles in a certain sequence, we induced multipolar magnetic dark modes and couple of molecular CTP shoulders along deep-UV spectra due to redistribution of charges excited from the conductive nanofilm.

2. Numerical modeling

We carried out the finite-difference time-domain (FDTD) method to predict and extract the optical properties of the proposed nanostructures, and also to monitor the plasmonic resonant bright and dark modes and their contribution in formation of antisymmetric Fano dip along the deep-UV band. In the numerical analysis, to determine the plasmonic responses, following parameters were employed: The spatial grid sizes in all three axes were set to $d_x = d_y = d_z = 1$ nm, and 48 perfectly matched layers (PMLs) were the boundaries for the workplace. Additionally, simulation time step was set to the 0.01 fs according to the Courant stability. The light source was a linear plane wave electric source with a pulse length of 2.6533 fs, offset time of 7.5231 fs.

3. Results and discussion

Here, firstly, we begin by examining the probability of inducing CTPs in both conductive film-mediated symmetric and

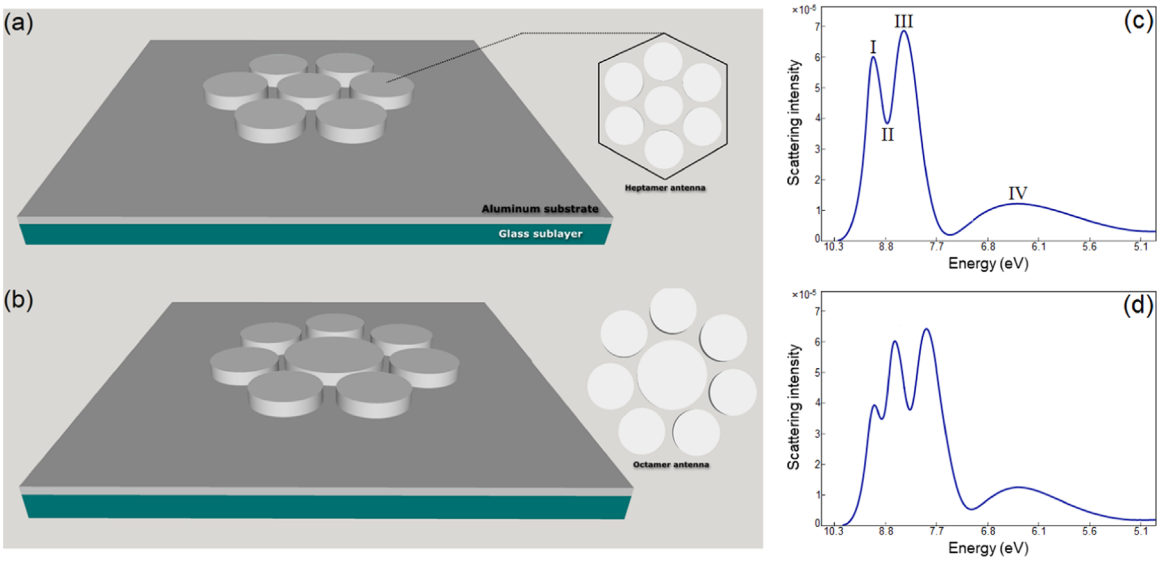

Fig. 1. (a, b) Perspective schematics for the symmetric heptamer and antisymmetric octamer antennas deposited on a conductive aluminum substrate, (c, d) the scattering intensity profiles for both aluminum heptamer and octamer antennas on a conductive substrate, indicating four different spectral features.