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Beyond dipolar regime in high-order plasmon mode bowtie antennas

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ABSTRACT

Optical nanoantennas have shown their great potential for far-field to near-field coupling and for light confinement in subwavelength volumes. Here, we report on a multimodal configuration for bright and polarization-dependent bowtie antenna based on large and highly crystalline gold prisms. Each individual prism constituting an antenna arm sustains high order plasmon modes in the visible and near infrared range that allow for high field confinement and two-dimensional optical information propagation. We demonstrate by scanning two-photon luminescence (TPL) microscopy and numerical simulations based on the Green dyadic method that these bowtie antennas result in intense hot spots in different antenna locations as a function of the incident polarization. Finally, we quantify the local field enhancement above the antennas by computing the normalized total decay rate of a molecular system placed in the near field of the antenna gap as a function of the dipole orientation. We demonstrate the existence of a subtle relation between antenna geometry, polarization dependence and field enhancement. These new multimodal optical antennas are excellent far field to near field converter and they open the door for new strategies in the design of coplanar optical components for a wide range of applications including sensing, energy conversion or integrated information processing.

1. Introduction

The development of experimental methods for optical imaging of single molecules and nanoparticles [1–4] has opened the doors to new branches of Optics where optical and plasmonic studies can be performed at the single object scale in confined geometries. Recent breakthroughs have highlighted the necessity to optically address and control localized or molecular light sources at the nanoscale in complex environments [5,6]. However, the mismatch between the absorption cross-section of molecular emitters and the effective wavelength of an incident electromagnetic wave is a recurrent issue for efficient coupling. Generally, an antenna converts propagative waves into a localized source of energy. The plasmonic version of an antenna establishes a bridge between far-field photonic modes and highly enhanced electric field hotspots that allows to overcome the aforementioned limitation [7–9].

Delocalized resonances on structured metallic films have shown a genuine potential for antenna applications [10–12], however energy conversion and enhancement effects have predominantly been observed with localized surface plasmon resonances (LSP) [8,9] and notably with subwavelength bowtie geometries [13–16]. Spectacular results have been obtained in several fields in optics like metasurfaces [17,18], non-linear optics [19,20], Raman spectroscopy [21], enhanced absorption in semiconductors [22] or fluorescence directivity control [23]. Plasmonic antennas based on LSP do not offer the same versatility than their electronic counterpart. Indeed, when coupled to microelectronic devices, an antenna not only concentrates the electromagnetic field in a small region of space, but also processes information by connecting the incoming signal to circuits and logical components. In order to transpose this concept to a plasmonic antenna, new designs and dimensions are to be considered to locally enhance the field and convey surface plasmon (SP) over distances larger than the SP effective wavelength too.

Mesoscopic colloidal gold platelets sustain high order SP modes in the visible and near infrared [24,25] and combine the features of delocalized and localized resonances that foster optical information transfer (at two dimensions) and subwavelength localization of the electric field in the same structure. Interestingly, these ultra thin plasmonic cavities have also been suggested as building blocks to develop 2D modal logical components [26]. They are therefore good candidates for bridging the concepts of plasmonic antennas and plasmonic circuits for integrated optical information processing.

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In this paper, we demonstrate experimentally that two prismatic plasmonic cavities coupled in a symmetric (both arms with similar sizes) or asymmetric (each arm has a different size) bowtie configuration exhibit a polarization-dependent antenna behavior. This new class of multimodal bowtie antennas offers a richer behavior than the common dipolar antennas and turns out to be a good input-output gateway from open space into nanoscale optical volumes. Their optical properties, probed here by two photon luminescence microscopy (TPL), originates from the specific spatial distribution of the high order modes sustained by each arm of the system [26]. The polarization dependent TPL maps are systematically compared to theoretical simulations based on a Green dyadic numerical tool that allows constructing TPL images by scanning a virtual Gaussian spot on the meshed metallic system [27]. In order to quantify the efficiency of these high order SP mode bowtie antennas, we compute the normalized total decay rate of a molecular system in the vicinity of the central hot spot as a function of the wavelength. We evidence two enhancement regimes that are directly driven by the multiscale nature of the coupled colloidal system.

2. Results and discussion

2.1. TPL investigation of the antenna modal structure

The prismatic structures (Fig. 1(a,b)) are obtained from a suspension of crystalline gold nanoprisms (see references: [24–26]) produced by reduction of Au precursors by polyvinylpyrrolidone (PVP) in alkaline conditions through a one–pot protocol at room temperature. These plasmonic cavities exhibit a high aspect ratio with a thickness 20 ± 3 nm and lateral dimensions ranging from 0.5 to 1 µm. Nanoprisms of such dimensions bear high order plasmon modes mainly localized along the platelet edges ([26,28]). A droplet of the solution is deposited on 10 nm ITO covered glass substrates. The occasional and serendipitous assembly of two colloids leads to a variety of dimers that are carefully characterized and selected by scanning electron microscopy (SEM) imaging (Fig. 1(b)). Thanks to the homogeneity of the suspension, one often observes bowtie arrangements of two prisms. The slight shift and overlap of the two apexes at the antenna center shown in Fig. 1(b) is a direct consequence of this complex fluidic assembling process.

We have used a home–built TPL optical microscope coupled with a tunable Ti:Sapphire femtosecond laser delivering 120 fs linearly polarized near–infrared pulses (Fig. 1(c)) [26]. The whole set of experiments presented here has been carried out at 700 nm. Antennas and the supporting ITO–covered glass substrates are placed on a XY piezostage and scanned in the excitation focal spot (300 nm FWHM) pixel by pixel for image reconstruction.

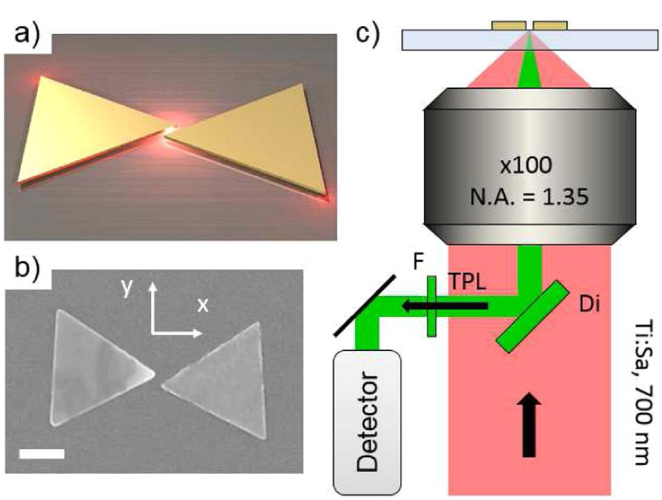

Fig. 1. (a) Artistic view of a multimodal bowtie antenna composed of two prismatic plasmonic cavities. (b) SEM image of a symmetric bowtie antenna made of two pristine colloidal gold prisms. scale bar represents 200 nm. (c) Schematic drawing of our home made microscope in which a femtosecond laser beam tuned at λ_L = 700 nm has been used for the experiments.

Two typical TPL maps recorded on a symmetric antenna with a gap of 7 nm are presented in Fig. 2(c,d) for two orthogonal polarizations. Generally, in standard subwavelength antennas, the strongest response is obtained for a polarization parallel to the main axis of the structure while a 90° rotation of the polarization mutes the TPL signal ([29,30]). Interestingly, the high order SP mode antenna shows a completely different behavior. It exhibits a strong TPL response at the central gap position for a polarization at 90° (perpendicular to the main axis x axis). With a polarization at 0° now parallel to the main axis, two bright TPL spots are recorded at two remote and symmetric locations labeled A and A′. These two hotspots are less intense than the central one and are separated by a distance of almost 900 nm that is larger than the effective SP wavelength on a flat gold film (λ_{SP} = 679 nm).

When we consider the TPL dynamics of the individual prisms that compose each arm, we observe differences compared to non-coupled individual objects. Isolated nanoprisms exhibit a sequential lightning of the apexes as a function of the rotating linear polarization (C_{3v} TPL symmetry reported in [31]). Indeed, when TPL images recorded on a prism for two orthogonal polarizations are superimposed, we obtain three equivalent hotspots located atop each apex that match the underlying symmetry of the prismatic colloid. Applying this methodology to images (c) and (d) in Fig. 2 for the dimer of nanoprisms shows that the trigonal symmetry of each arm is not preserved since the positions C and C′ are mute. In addition, the intensity in B-B′ and A-A′ are not equivalent which confirms similar observations in a previous work [26]. The results show that this pair of cavities not only concentrates the electric field in a specific region of space (similar to lightning rod effect), but also forms a coupled system.

Since TPL is related to the field enhancement in the metal, this microscopy is used for near–field mapping in plasmonic systems [32]. It also has been recently demonstrated that the TPL signal is not only restricted to field mapping but results from the convolution of the squared in-plane SP-LDOS (Surface Plasmon Local Density of States) with the Gaussian profile of the excitation light beam [26].

$$I_{TPL}(\mathbf{R_0}, \omega) \propto \omega^4 \int_V |\mathbf{E_0}(\mathbf{R_0}, \mathbf{r}, \omega)|^4 \rho_{\parallel}^2(\mathbf{r}, \omega) d\mathbf{r} \quad (1)$$

Where $\mathbf{E_0}$ is the laser electric field impinging at the position $\mathbf{R_0}$. ω is the incident frequency and V is the volume of the metallic particle. $\rho_{\parallel}^2$ corresponds to the squared in-plane total density of states within the metal accounting for the exact geometry of the system and the surrounding properties. The details of the Green dyadic formalism used in this work are described in the references: [33,26]. TPL microscopy can therefore be used to get information on both field enhancement and modal distribution [30,31]. The combination of TPL mapping and simulations provides a convenient tool for the assessment of the optical properties of these multimodal bowtie antennas in complement to electron [34] or fluorescence based methods [35,36].

From the SPLDOS computation, we deduce that each 450 nm long arm of the antenna in Fig. 2 sustains a m=3 SP mode that displays nodes and antinodes along the edges. High order modes borne by extended plasmonic resonators have shown to be robust when the metallic structure undergoes moderate modifications [37]. However either a significant or a moderate spatial redistribution of the SP modes can be achieved by inserting a defect in the platelet [31] or by electromagnetically coupling two cavities [26]. The latter case is illustrated in Fig. 2 and confirmed by the SPLDOS computation shown in Fig. 2(b). The two m=3 mode distributions are moderately impacted by the presence of the opposite structure because of the small interaction area delimited by the facing metallic surfaces in the gap. Yet, the experimental TPL response of the nanoprisms is dramatically