Glimpsing non-radiative plasmonic dark modes in the near field: exciting the unexcitable

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Abstract

We present near-field mapping of a system of coupled nanoantennas composed of bright and dark plasmonic modes. We identify the governing geometrical and illumination parameters that affect the *excitation* and the *structure* of the induced dark mode. Based on the insights we obtain in this study, we demonstrate, in the nearfield, how the destructive interference in a plasmonic system occurs that causes EIT-like effect. Our findings have potentials in the fundamental study of excitation mechanism in plasmonic nanostructures and will serve as a design guideline for plasmonic sensors.

1. Introduction

Near-field interactions among plasmonic entities have been extensively studied in the past decades leading to a myriad of useful properties with useful, real-world applications such as enhanced Raman scattering, spectral modulation in plasmonic sensors and sub-wavelength nanolithography. In its simplest form, plasmonic interaction between two resonant structures results in a coupled system that can be conveniently described using coupled oscillator models. Successful application of such models have been demonstrated in plasmonic systems exhibiting Fano resonances.[1-2]

Of particular interest are resonator systems that exhibit particularly balanced coupling of one "bright" resonator mode (dipole-allowed, directly excitable by external radiation) and one "dark" resonator, usually not directly excitable by external radiation fields of dipolar symmetry; only by a gentle break in the overall symmetry of the coupled system allows to excite such modes. These coupled systems offer unique opportunities for the study of resonant modes not otherwise accessible. In the domain of metamaterials, such coupling has been exploited to observe the plasmonic version of Electromagnetically-induced transparency (EIT) phenomena.[3-4]

Although this phenomenon is a direct result of strong near-field coupling, experimental observation is usually made in the far-field and is assigned to symmetry breaking that allows the excitation of the dark modes. Here, we investigate using a system of two metallic nanoantennas the phenomena of EIT directly in the nearfield and unravel the peculiarities of the coupling.

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2. Samples and methods

The structures are planar and made up of three gold wire-like nanoantennas, arranged such that they form an "H" shape as shown in Fig. 1. It has to be stressed that all entities forming the "H" are separated, hence conductive coupling among them is excluded. In what follows, we name our structures as *H* or *I* depending on the polarization of the exciting field: the *H* structure corresponds to the incident electric field that is polarized parallel to the central nanoantenna connecting the two side arms; the *I* structure, the incident electric field is polarized parallel to the two side arms.

The structures are defined on silicon dioxide substrate using electron beam lithography, followed by thermal evaporation of gold and a lift-off process to remove the polymer template. Geometrical parameters are indicated in Fig. 1a. A set of samples made from a combination of two lengths of wires, $l_{1,2}$, were fabricated. The wire lengths were chosen to be either 140 nm or 520 nm long that support, respectively, the 1st order and the 3rd order resonance. [7]

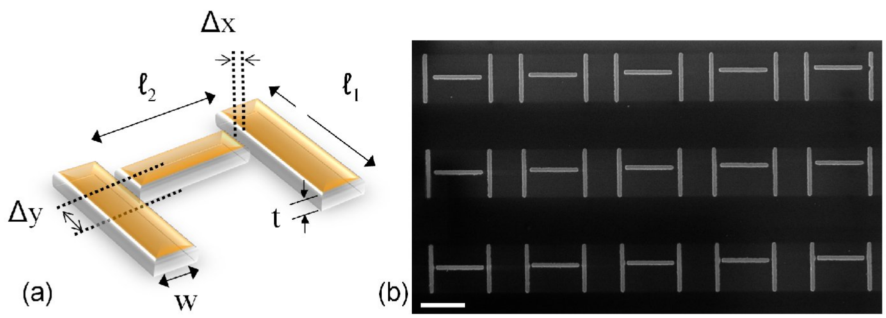

Fig. 1: Structure parameter of the experimental structures. (a) Schematic of structural dimension.
(b) An SEM image of the sample made of 3rd order resonance nanowires. The white bar is 500 nm long.

All the wires have the same width w (40nm) and thickness t (30nm). Using a combinatorial approach, we fabricated structures with varying coupling distance (Δx) between the center wire and the side arms and with varying vertical displacement (Δy). Figure 1b shows an SEM micrograph of a fabricated sample composed of wires sustaining the 3rd order resonance in the near-infrared. In the image, Δx increases systematically from the bottom to the top row, whereas Δy increases systematically from left to right.

The optical experiments were carried out using a home-built apertureless scanning near-field microscope (aS-NOM)[5-6] and compared to numerical simulations using a Finite-Difference Time-Domain (FDTD) method.

3. Results and discussion

The combinatorial approach allows us not only to directly determine the most suitable geometries for optimally resonant (bright) nanoantennas,[7] but also to study the influence of the relative arrangement between nanoantennas. In particular, we find clear evidence for the possibility of strong, near-field coupling induced excitation of "dark" nanoantennas – i.e., isolated antennas that are hardly excitable at all, the oscillation of which, however, can be induced in the presence of a "bright" twinned nanoantenna. We show that the mode structures and spatial mode overlap between the bright and dark antennas are crucial factors that govern the behavior of the system. We also show that there exists a strong dependency on excitation symmetry and polarization. Figure 2a shows experimental near-field amplitude of the z component of electric field on the *H* structure shown in Fig. 1b. It is seen that the ampli-

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tude of the bright ‘horizontal’ bar and the dark ‘vertical’ bars are varied depending on the values of Δx and Δy. Most notably, it is witnessed that the dark eigenmode is excited with a noticeable strength (see vertical nanowires row 4, column 4-6). That is possible in a geometrical configuration where the field distribution of the bright mode is matched to that of the dark mode to allow for an efficient energy transfer. Experimental results are in good agreement with the FDTD simulations.

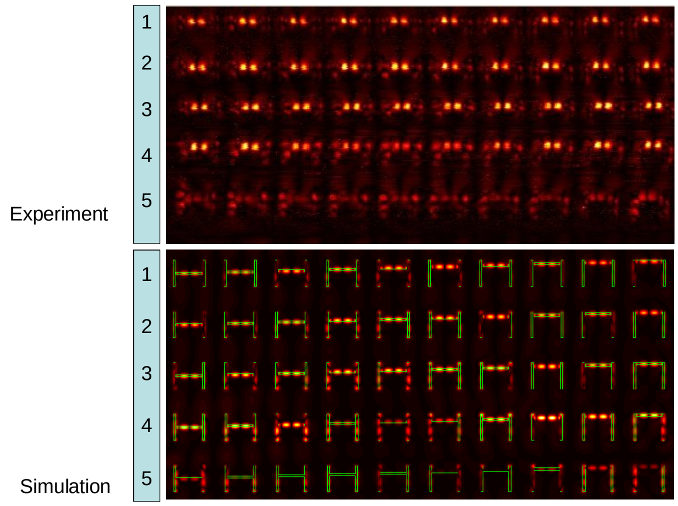

Fig. 2: Near-field optical amplitude. (a) aSNOM (b) FDTD simulation of the sample in Fig. 1b (H structure).

4. Conclusion

We present near-field imaging of non-radiative dark modes and show how they can be excited in the presence bright modes. Several parameters are identified for the tuning of the behavior of the systems, including for instance *coupling distance*, *symmetry breaking*, *polarization* and the *phase* of the incident light, as well as the orientation and the configuration of the sample. We wish to stress that the work contributes to the exploration of more complex excitation scheme of plasmonic resonances. As we show conclusively, it is possible to force the excitation of a highly structured bright mode by an otherwise extremely homogenous external illumination and this highly structured near-field in turn allows the near-field excitation of plasmonic modes otherwise inaccessible with the external illumination. Such kind of cascaded near-field excitation schemes promise to be of use to control the near-field inaccessible by means of far-field optics.

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