Symmetry breaking and nonlinear effects in PIT metamaterials

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Abstract

Second and third harmonic generations were experimentally investigated in plasmon induced transparency (PIT) metamaterials with unit-cells consisting of coupled radiative bright and non-radiative dark plasmonic elements. We will discuss these nonlinear phenomena and in particular their correlation to the effect of symmetry breaking and field enhancement.

A plasmonic mode in a metallic nanostructure can be either superradiant (bright mode) or subradiant (dark mode) depending on how efficient it can couple with incident light from free space. In particular, the coupling efficiency is closely correlated to the symmetry of the plasmonic mode and the polarization of the incident light field. Each meta-molecule consists of a single metal bar which interacts strongly with the incident light due to the excitation of an electric dipole mode and a pair of metal bars with perpendicular orientation (see Fig. 1A). The double bars possess an asymmetric mode that at normal incidence can only be excited by the near field coupling to the single metal bar (electric dipole mode). This asymmetric plasmon mode is characterized by current flow in opposite directions in the two bars which can be described by the sum of a magnetic dipole and an electric quadrupole [1].

The close proximity of the dark and bright element can lead to a strong coupling among them resulting in a pronounced hybridization of the Plasmon modes between the bright and the dark element. In such a configuration similar effects as atomic electromagnetic induced transparency (EIT) can occur [1]. However, the effect is based on classical destructive interference of Plasmon modes and therefore called Plasmon-induced transparency (PIT). Several slightly different designs were demonstrated to show such Plasmon-induced transparency behaviour [2-4]. Figure 1E shows the transmission spectra for a periodic array of the meta-molecules when the gap size between the single and double bar elements is increased (compare Fig. 1A-D). The transparency window (marked by II in Fig. 1E) is a result of destructive interference suppressing the absorption of the dipole mode.

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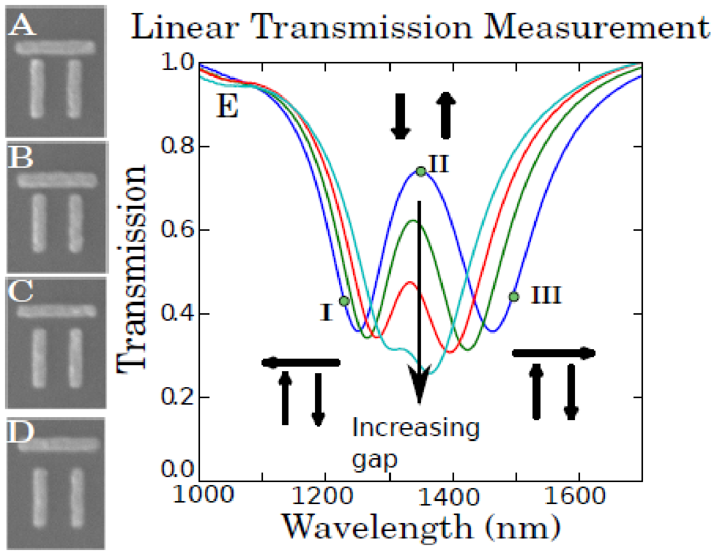

Fig. 1: (A-D) Scanning electron microscope images and transmission spectra (E) of the ├ shaped meta-molecules for different gap sizes. The period is 600 nm in both directions and the array size was 50x50 um^2 . Vertical bar size is 262x46 nm^2 with a 93 nm distance between bars. Horizontal bar size is 305x60 nm^2. The gold thickness is 35 nm. Structures with the following gaps between the horizontal and vertical bars were fabricated: A) 20 nm gap; B) 35 nm gap; C) 50 nm gap; D) 80 nm gap. The arrows in E represent the direction of the displacement current at the wavelengths indicated by I, II, and III.

In atomic EIT systems, the nonlinear properties of the medium are strongly enhanced at the transparency band. In particular, the increase in the group index of the medium together with high transmission and low absorption loss lead to interesting third-order nonlinear effects. These effects should also exist in the PIT system. More interestingly, the PIT structure shows a strongly broken inversion symmetry and should also give rise to a non-zero second harmonic generation signal. In particular, the distance between the bright and dark element influences the coupling strength and therefore determines how strong the inversion of the total element is broken, whereas the third harmonic signal should mainly depend on the field localization and field enhancement.

For our investigation of the nonlinear signal from a periodic pattern of PIT elements we follow a numerical approach introduced by Zeng *et al.* [5]. Our theoretical studies are supported by experimental observations of the spectrally resolved nonlinear optical signal for different gap sizes of the structure. By tuning a short laser pulse over the entire near-infrared spectral range we determine the signal strength for the second and third harmonic light.

To characterize the nonlinearities of PIT meta-molecule we measured the visible light emission with a spectrometer while illuminating with 200 femto-second pulsed infrared light from a synchronously pumped optical parametric oscillator. Both the second and third harmonic emission are observed. The detailed findings on the dependence of the nonlinear generations on the coupling will be presented in the conference.

Plasmonic-induced transparency can provide a novel approach for tailoring the optical properties of plasmonic metalmaterials. Especially, the increased group index could be useful for nanophotonics applications. Here, we showed that also the symmetry of the unit-cell and the coupling between different Plasmon modes can give rise to a distinct behaviour in the nonlinear response of the structures.

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