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Influencing the ultrafast plasmon damping time using Fano resonances for nonlinear plasmonics

Krishnan Thyagarajan[1] and Olivier J. F. Martin[1]

[1]Nanophotonics and Metrology Laboratory (NAM), Swiss Federal Institute of Technology Lausanne (EPFL), 1015 Lausanne, Switzerland

Abstract. We explore the possibility of strongly influencing the plasmon damping time in nanostructures for efficient second harmonic generation, by taking advantage of the tunability of the narrow linewidth feature in the extinction cross-section exhibited by plasmonic nanostructures under Fano resonance.

1. Introduction

With the ability to generate extremely strong and confined optical fields at the nanoscale, resonant metallic nanostructures supporting localized surface plasmon modes play a crucial role in current nanoscience. Such strong confined fields allow one to use these structures to study and control nonlinear optical (NLO) signal generation at subwavelength scales. Second harmonic generation (SHG) is one such important nonlinear optical effect which has the added advantage of being sensitive to symmetry. In earlier works, we proposed and tested the possibility to enhance SHG from nanostructures using Fano resonances [1], or double-resonant plasmonic antennae [2]. In this work, we propose to study the influence of the plasmon damping time – both radiative and non-radiative – at the fundamental frequency on the intensity enhancement at the second harmonic.

2. Theoretical background

SHG from centrosymmetric nanostructures originating from the breaking of inversion symmetry at their surfaces is a well-known phenomenon [3]. Yet, there is always a need to make the process more efficient, by reducing unwanted losses. Many recent developments have strived to reduce the plasmon damping in metallic nanostructures. Recently, nanostructures such as plasmonic oligomers have been studied and have shown strong hybridization of their constituent resonant primitive plasmonic modes, leading to new hybridized superradiant ‘bright’ and subradiant ‘dark’ modes (see Fig. 1a). In such systems, the incident photons excite the bright mode, which couples to the dark mode via both a spectral and spatial overlap, and the interference of both of these modes leads to the characteristic Fano resonance.

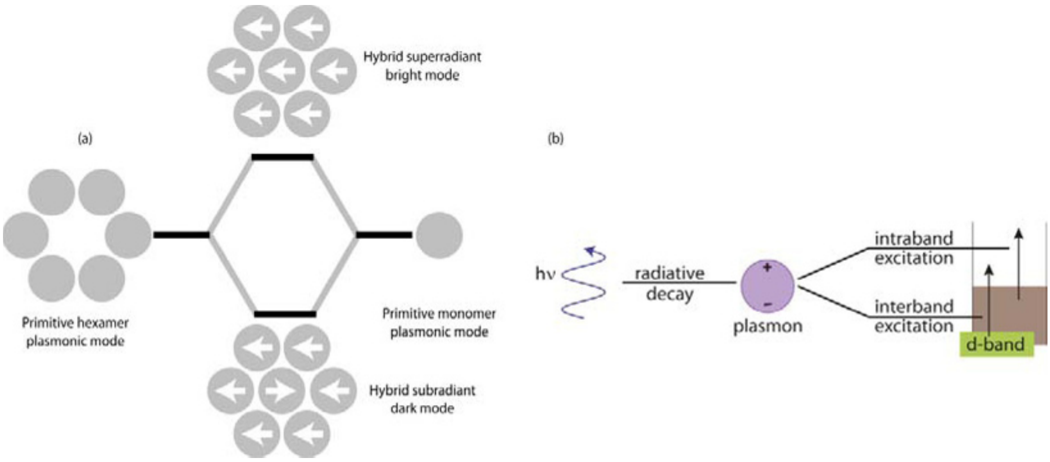

Fig. 1: (a) Plasmon hybridization scheme showing the superradiant 'bright' and subradiant 'dark' plasmonic modes that are important for the Fano resonance to occur (b) Schematic diagram showing the radiative and non-radiative loss mechanisms in plasmonic nanostructures

Losses in metallic nanostructures, which can be of two kinds – *radiative* and *non-radiative* (see Fig. 1b) – can be accounted for by using an equivalent complex resonance frequency; the real part of which corresponds to the mode resonance frequency ω_d while the imaginary part γ_d accounts for the intrinsic damping. For the dark mode, which does not couple directly to the incoming field, there are two possible channels for losses, *non-radiative* losses which involve the dissipation of energy as heat, and '*radiative'* losses, which can be thought of as the result of the coupling of the dark mode to the bright mode. The latter, which can then radiate this energy out, gives this second loss mechanism of the dark mode the name of '*radiative*' losses. It is this latter loss that causes the Fano dip to widen.

As discussed in an earlier work [1], for the case of SHG, it is desired that the structure is engineered such that the fundamental frequency of the pump laser lies at a minimum of the scattering cross section of the structure, so that a lesser fraction of the fundamental frequency field undergoes radiative losses and a greater fraction can be converted to the second harmonic. In this work, we show the importance of controlling these two loss channels of the dark mode to efficiently enhance the near field intensity. We engineer such a condition using plasmonic oligomers made of silver for optimized SHG in the far-field. By engineering these structures, we are able to change the plasmon damping lifetime and therefore also the radiative lifetimes, which in turn should influence the SHG from plasmonic nanostructures.

3. Simulations

Plasmonic heptamers consist of seven individual cylinders arranged into a symmetric structure with six of them forming an outer ring and the seventh being present at the center of the ring. The structure supports a multipolar-like dark mode and a strong dipolar bright mode which together give rise to the Fano lineshape in the extinction cross-section. The near field profile of these structures can be modified by changing their geometrical parameters such as the gap between the structures, the diameter of the individual structures, etc. The stimulated structures have a 150 nm diameter , a 40 nm height and a gap of about 30 nm in between them. As can be seen in Fig. 2, the Fano lineshape can be tuned to exhibit a strong dip in the case of strong coupling or a weak dip in the case of weak coupling. It is possible to show that the ideal regime where the intensity enhancement is the strongest while maintaining low radiative and non-radiative losses occurs when the structures are fabricated such that the $\gamma_{rad} = \gamma_{non-rad}$, that is, the radiative and non-radiative losses are equal [4]. It is this regime that we are interested to work with, to maximize the SHG in the far-field.

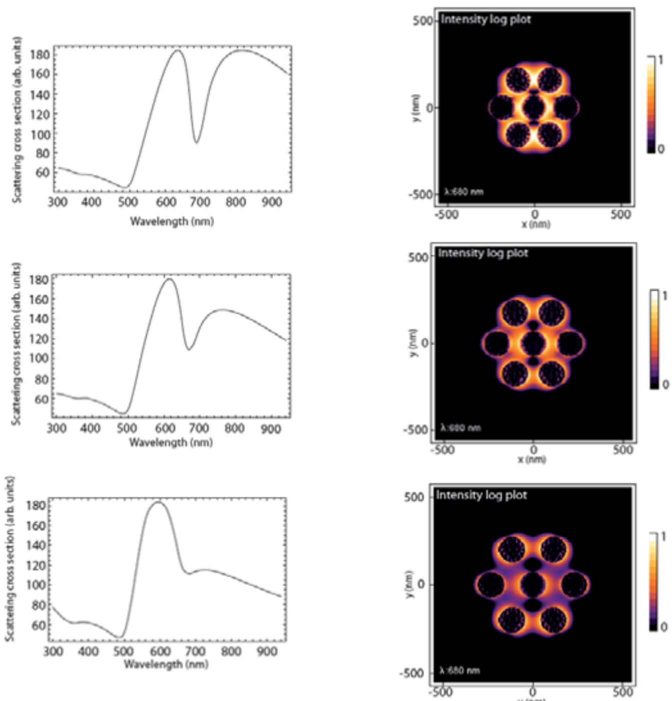

Fig. 2: Simulations showing the scattering cross-section and the near-field profiles for the different configurations of the heptamers exhibiting Fano resonances.

4. Conclusions

It was seen that the coupling between the two modes – the bright and the dark – increases as the gap between the structures decreases, thereby increasing the lifetime of the energy stored in the region. But simultaneously there is a trade-off for as the energy stored in the gap increases, so do the losses. Therefore there is an optimum configuration that allows maximal intensity enhancement in the gap, which will therefore correspondingly increase the second harmonic signal generated from the structures. The experimental realization of this is currently in progress.

5. References

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