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Tailoring the field enhancement in Fano-resonant nanoantennas for improved optical bistability

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Abstract. The influence of Fano resonances on the nonlinear response of hybrid plasmonic nanostructures, i.e., nanoantennas loaded with a nonlinear optical material, is theoretically investigated using the combination of a surface integral equation method and an analytical model. The results demonstrate that a suitable design of the field enhancement enables the observation of optical bistability for incident conditions that would be impossible for a bare nanoantenna. This study provides new insights into the possibilities offered by Fano resonances to control the nonlinear response of hybrid plasmonic systems. © *2017 Society of Photo-Optical Instrumentation Engineers (SPIE)* [DOI: 10.1117/1.JNP.11.016007]

Keywords: optical bistability; nanoantenna; Kerr effect; nonlinear plasmonics; surface integral equation method.

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1 Introduction

Plasmonic nanostructures have the ability to concentrate light down to the nanoscale, far below the diffraction limit.[1] The high electric field associated with localized surface plasmon resonances (LSPRs) enables the observation of nonlinear optical processes, such as second harmonic generation,[2] third harmonic generation,[3] multiphoton photoluminescence,[4,5] and Kerr effect.[6] More recently, the nonlinear optical responses of hybrid plasmonic nanostructures have been intensively investigated, revealing new and interesting features.[7] In particular, optical bistability in nonlinear metal nanostructures interacting with Kerr materials has been predicted and experimentally observed.[8] Several configurations have been considered, including loaded nanoantennas,[9] subwavelength slit apertures,[10,11] metal–dielectric nanoparticle dimmers,[12] plasmonic nanoshells loaded with a nonlinear medium[13] following a design first proposed for enhanced second harmonic generation,[14] metallic nanoparticles coated with nonlinear shells,[15] and film-coupled plasmonic nanocubes.[16] The influence of nonlinearity on the propagation of surface plasmon polaritons at metal/dielectric interfaces has been investigated.[17,18] In the case of plasmonic nanoantennas loaded with a Kerr medium, an analytical model was developed by Zhou et al.[19] This model predicts the influence on the optical bistability of the different LSPR properties (namely the width, the amplitude of the field enhancement, and the amplitude of the resonance shift as the refractive index of the load varies). In summary, the observation of optical bistability is possible at low pump intensity provided that there is[19]

- a strong field enhancement in the nanoantenna gap, which results in a strong intensity in the nanoantenna load;
- a narrow LSPR (note that the LSPR spectral width is directly related to the total losses in the nanosystem);
- a large spectral shift of the LSPR for a given variation of the refractive index of the load.

Different approaches have been proposed for optimizing these LSPR properties, including the introduction of a gain medium[20] and the control of the radiative properties with a reflecting

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metallic surface.[21] Unfortunately, a general and unique method for the control of these three parameters at the same time is still missing.

Fano resonances have proved their ability to tailor the spectral responses of plasmonic nanosystems.[22,23] Indeed, Fano resonances arise from the interference between a radiative mode and a second mode, which can be radiative or not. They are characterized by an asymmetric line shape, as opposed to their Lorentzian counterparts observed in general in the field of plasmonics. Fano resonances have been observed in plasmonic nanostructures with various geometries, and several theoretical methods have been developed to understand and predict their behavior.[24,25] At the same time, the field distribution close to Fano-resonant plasmonic nanostructures is also strongly dispersive.[26–28] For instance, Fano resonances have been used in nonlinear plasmonics, mainly for inducing a stronger field enhancement at the pump wavelength.[29–31] However, these previous studies exploited only the stronger near-field intensity induced by Fano resonances but did not take advantage of the modification of the other parameters, i.e., the resonance width and shift. Recently, the influence of the nonlocality on Fano resonances and the enhancement of the optical bistability in coated cylinders have been theoretically investigated.[32]

In this article, we demonstrate that Fano resonances can control the three LSPR parameters (field enhancement, resonance width, and spectral shift) that are instrumental to the observation of optical bistability.[19] Starting with a plasmonic nanoantenna loaded with a nonlinear Kerr medium, we add two gold nanorods in its vicinity to generate a Fano resonance.[33] We show that the resonant enhancement of the field at the center of the nanoload depends on the nanorod length and that the field enhancement, the resonance width, and the spectral shift can be simultaneously improved for a specific configuration.

2 Results and Discussion

The article is organized as follows. First, the linear optical responses of both the nanoantenna and the Fano-resonant nanostructure are evaluated using the surface integral equation method.[34] The influence of the Fano resonance on the field enhancement is discussed in detail. In the second part, the nonlinear response resulting from the Kerr effect occurring in the load of the dipolar nanoantenna and Fano-resonant nanostructure is investigated using an analytical model adapted from Ref. 19.

2.1 *Linear Responses of the Nanoantenna and Fano-Resonant Nanostructure*

The gold nanoantenna studied in this work is shown in Fig. 1(a). The nanoatenna is composed of two 70 nm nanorods separated by a gap $g = 25$ nm. The width and height of the nanorods are 40 nm. The polystyrene load is shown in blue, and the gold parts are shown in yellow. The refractive index of the surrounding medium is 1.33, and the linear refractive index of the polystyrene load is $n_0 = 1.60$. The optical response of the nanoantenna has been computed using a surface integral equation method with a typical mesh side of 7 nm.[34] The nanostructure is driven by an incident planewave propagating perpendicularly to the structure and polarized along the nanorods long axis [Fig. 1(a)]. The intensity close to the nanoantenna is shown in Fig. 1(b) for an incident wavelength $\lambda = 730$ nm. It is clearly observed that the intensity is maximal in the nanogap and that the intensity is almost homogeneous between the two nanoantenna arms. The backscattering and the enhancement factor for this nanoantenna are shown as functions of the incident wavelength as dashed red lines in Figs. 2(a) and 2(b), respectively. The enhancement factor is defined as the ratio between the intensity at the gap center and the incident intensity. As expected, a strong enhancement of the back scattering and of the intensity in the nanogap is observed for incident wavelengths close to the resonance wavelength.[34]

To investigate the influence of a Fano resonance on the intensity enhancement in the nanogap, two gold nanorods parallel to the nanoantenna have been added to the nanostructure [Fig. 1(a)].[33] The numerical simulation shows that the intensity distribution in the nanogap is also homogeneous in this configuration [Fig. 1(c)]. The backscattering has been evaluated for several rod lengths ($L = 80$ nm, 120 nm, and 160 nm), revealing Fano resonances with different

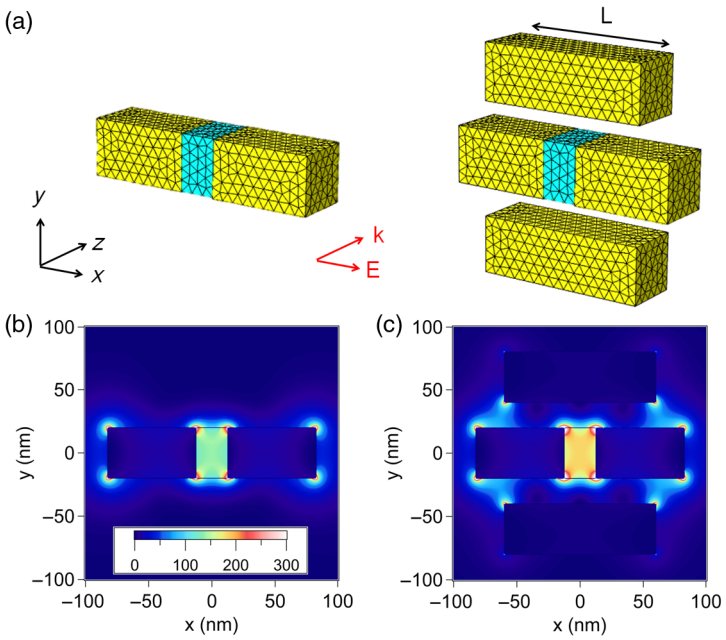

Fig. 1 (a) Surface meshes used for the numerical computations. The nanoatenna is composed of two 70-nm nanorods separated by a gap $g = 25$ nm. The width and height of the nanorods are 40 nm. The length of top and bottom rods is $L = 120$ nm. The nanostructure is driven by an incident planewave propagating perpendicularly to the structure and polarized along the nanorods long axis. The polystyrene load is shown in blue and the gold parts in yellow. Near-field intensity in the plane $z = 0$ close to (b) the nanoantenna ($\lambda = 730$ nm) and (c) the four-nanorod nanostructure ($\lambda = 750$ nm). The same color scale is used for the two figures.

lineshapes.[33] In these four-nanorod nanostructures, the Fano resonance arises from the coupling between the dipolar modes supported, respectively, by the nanoantenna and the two parallel nanorods, and their optical responses are well reproduced by an extended coupled oscillator model.[33] In this model, the coupled oscillators are driven by an external force with a harmonic time dependence representing the excitation by the incident planewave. The equations of motion for the two oscillators can be written as follows:[33]

$$\begin{cases} \ddot{x}_1 + \gamma_1 \dot{x}_1 + \omega_1^2 x_1 + g x_2 = 0.5\, P_{\text{tot}} + \alpha_1 E_{\text{ext}} \\ \ddot{x}_2 + \gamma_2 \dot{x}_2 + \omega_2^2 x_2 + g x_1 = 0.5\, P_{\text{tot}} + \alpha_2 E_{\text{ext}}\,, \end{cases} \tag{1}$$

where $\omega_{1,2}$ are the resonant frequencies, $\gamma_{1,2}$ are the damping constants, $x_{1,2}$ are the respective amplitudes, $\alpha_{1,2}$ are the polarizabilities, E_{ext} is the incident electric field, g is the constant of the coupling spring, and P_{tot} is the total dipole moment. As the length of the nanorods L increases, both the resonant frequencies of the mode supported by the two parallel nanorods and the mode coupling evolve, resulting in different Fano profiles.[24] This evolution is directly induced by the interaction between the dipolar mode supported by the nanoantenna and that supported by the two parallel nanorods. Indeed, in this configuration, the nanoantenna not only is driven by the incident field but also interacts with the two parallel nanorods. As a consequence of this mode coupling, the enhancement factor is expected to evolve as the nanorod length changes, i.e., as the Fano resonance evolves. The enhancement factor is shown as a function of the incident wavelength for several nanorod lengths L ranging from 80 to 160 nm [Fig. 2(b)]. For all the considered gold nanorod lengths L, a modification of the wavelength of the maximum enhancement factor, the maximum of the enhancement factor, and the width of the resonance are observed, demonstrating that these important parameters can be controlled by the Fano resonances. However, only a length $L = 120$ nm results in modifications beneficial for the observation of optical bistability. For $L = 80$ nm, the enhancement factor is lower than the one obtained for the isolated nanoantenna and, for $L = 160$ nm, the width is larger than the one

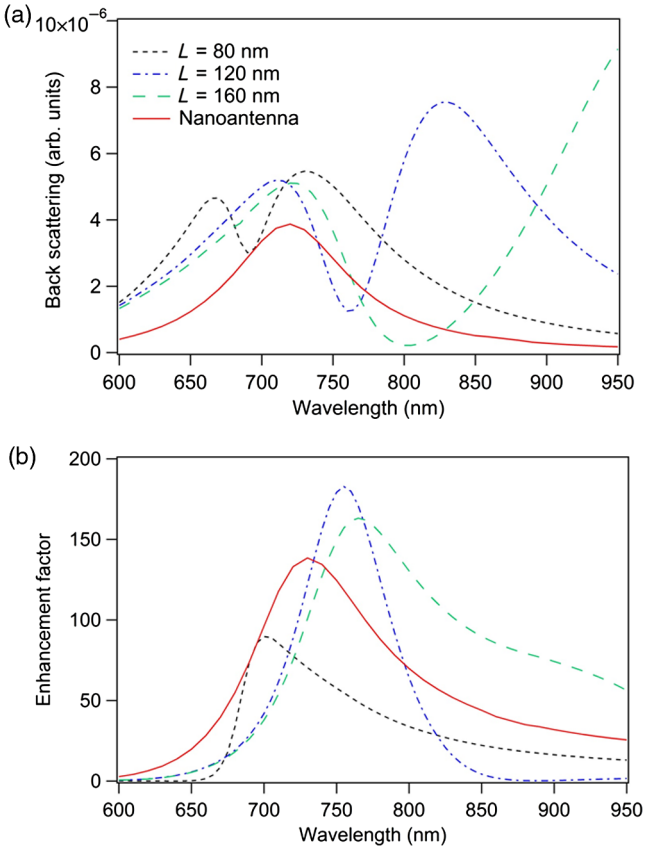

Fig. 2 (a) Backscattering and (b) enhancement factor as functions of the incident wavelength for an isolated nanoantenna (red dashed curves) and the four-nanorod nanostructure with $L = 80$ nm (black curves), 120 nm (blue curves), and 160 nm (green curves).

obtained for the isolated nanoantenna, since the full width at half maximum is increased by ~35%. For this reason, only the length $L = 120$ nm is considered in the following sections.

2.2 *Optical Bistability*

Now, let us consider the nonlinear Kerr effect in the polystyrene load present in the nanoantenna gap following the approach developed in Ref. 19 and extended in Ref. 21. The main approximation of this model is to consider that the electric field distribution is uniform in the nanogap, resulting in a uniform refractive index change. Figure 1(c) shows that this approximation is also valid in the case of the Fano-resonant nanostructure. Indeed, the intensity in the nanogap does not vary much over most of the load volume [Fig. 1(c)]. In this approach, the refractive index of the polystyrene becomes a function of the intensity: $n = n_0 + n_2 I$, where n_0 is the linear part of the refractive index, n_2 is the nonlinear Kerr coefficient, I is the intensity in the Kerr material, i.e., in the nanogap, and $I = 1/2\, c\, \varepsilon_0 |E|^2$.[35] Note that this approximation is correct for relatively small changes of the refractive index as those considered here.[9,19,21] The nonlinear Kerr coefficient of polystyrene is $n_2 = 1.14 \times 10^{-12}$ cm^2/W.[36] To determine the influence of an increase of the load refractive index on the optical response of the nanostructures, computations have been performed considering different values for this refractive index (Fig. 3). The simulations show that all the important parameters discussed in Sec. 1 are indeed improved in the Fano-resonant structure. The spectral shift is increased by ~15% [Fig. 3(a)]; the maximum of the enhancement factor is increased by ~50% [Fig. 3(b)]; and the width of the resonance is divided by approximately a factor 2 [Fig. 3(c)]. These results predict that the optical bistability in hybrid plasmonic structures can be actually improved due to the Fano resonance.

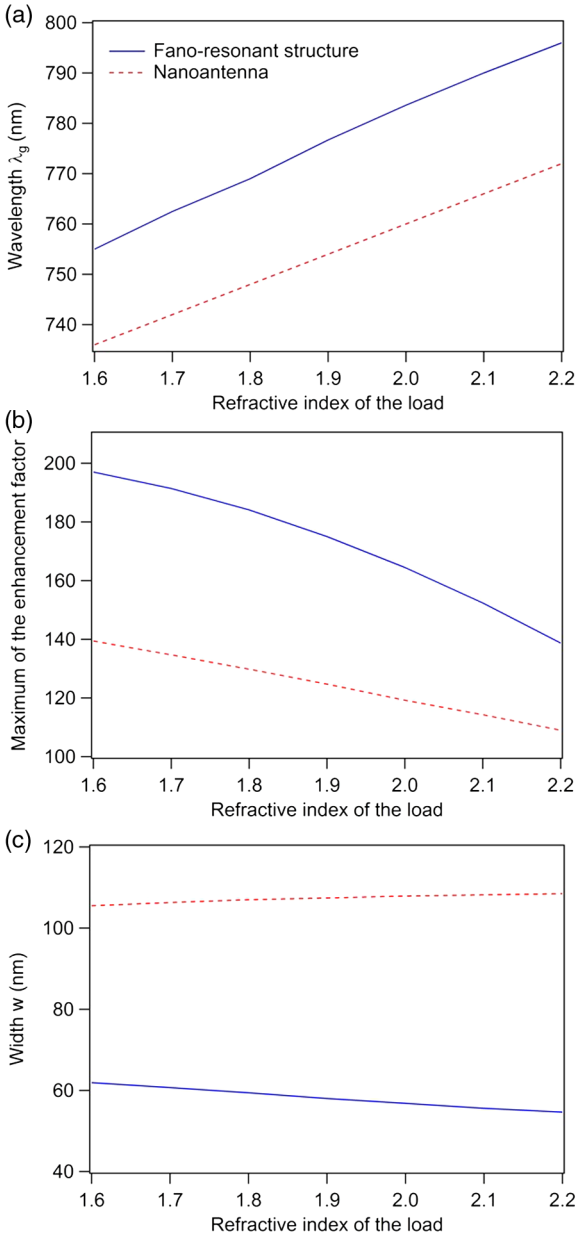

Fig. 3 (a) Wavelength of the maximum enhancement factor, (b) maximum of the enhancement factor, and (c) width *w* as functions of the refractive index of the load for the four-nanorod gold nanostructure (solid blue curves) and the nanoantenna (red dashed curves).

To unambiguously confirm the potential of the proposed approach, the nonlinear relation between the enhancement factor and the pump intensity has been determined for an incident wavelength $\lambda = 820$ nm using an analytical model [Fig. 4(a)]. The reader is referred to previous publications for the implementation details.[19,21] In the case of the Fano resonance, a hysteresis loop is clearly observed for incident intensities between 5.7 and 6 GW/cm^2, while no bistability is observed in the nanoantenna for any pump intensity. Actually, the analytical model proposed by Zhou et al. confirms that optical bistability cannot be observed in the nanoantenna for this incident wavelength.[19] However, bistability is possible at a slightly longer incident wavelength (825 nm). In this case, the threshold is 7.4 GW/cm^2. This value is ~50% higher than the one obtained for the Fano-resonant nanostructure, emphasizing the benefit of Fano resonances for the observation of optical bistability. On the other hand, it is difficult to assess experimentally the intensity in the nanogap, and far-field measurements are by far more convenient.

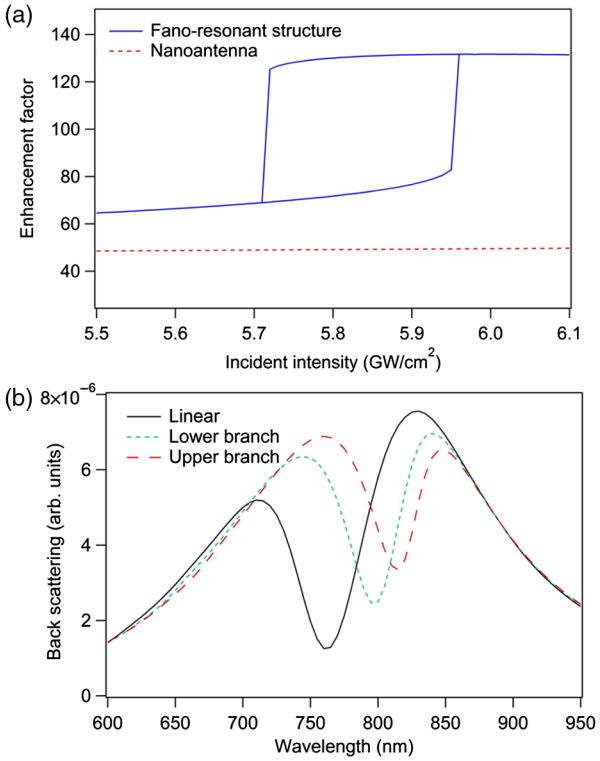

Fig. 4 (a) Enhancement factor as a function of the incident intensity for the four-nanorod gold nanostructure (full blue curve) and the nanoantenna (red dashed curve). The incident wavelength is 820 nm. (b) Backscattering as a function of the incident wavelength for linear case (black curve), the lower branch (green curve), and the upper branch (red curve). The nonlinear spectra are evaluated at 5.72 GW/cm^2.

The backscattering is shown in Fig. 4(b) as a function of the wavelength for the linear case ($n = n_0$), the lower branch ($n = 2.10$), and the upper branch ($n = 2.35$) for an incident intensity of 5.72 GW/cm^2 corresponding to the beginning of the bistable region. The lineshape of the Fano resonance evolves as the refractive index of the nanogap increases, and a modulation of the backscattering as high as 40% is observed at specific wavelengths between the two branches, underlying that the bistability is clearly observable in the far-field region. Note that a change of the load refractive index not only shifts the resonance as observed for a classical LSPR with a Lorentzian profile but the lineshape of the Fano resonance is also modified. This observation is a direct consequence of the relation between the nanoantenna properties and the mode coupling, as described by the extended coupled oscillator model in Eq. (1).

3 Conclusions

In summary, it was demonstrated that the Fano resonance provides control on the three LSPR parameters (its field enhancement, resonance width, and spectral shift) that are key for the observation of optical bistability in hybrid plasmonic nanostructures. Starting with a plasmonic nanoantenna loaded with a nonlinear Kerr medium, two gold nanorods were added in its vicinity to produce a Fano resonance. It was shown that the resonant enhancement of the field in the nanogap depends on the nanorod lengths, and the field enhancement, resonance width, and spectral shift can be simultaneously improved for a specific nanorod length. This improvement results in optical bistability for pump conditions impossible for a bare nanoantenna. The proposed method is very flexible and can be combined with other ones, such as the introduction of a gain medium[20] and the control of the radiative properties with a reflecting surface.[21] In addition, more efficient nonlinear materials, such as epsilon near-zero materials, could also be considered to load nanoantennas.[37,38] Fano-resonant nanostructures loaded with a Kerr medium represent very

promising building blocks for nonlinear metamaterials for efficient phase conjugation and for the design of a time-reversal "perfect lens."[39]

Acknowledgments

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