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Manipulating the Optical Bistability in a Nonlinear Plasmonic Nanoantenna Array with a Reflecting Surface

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Abstract The influence of a reflecting surface on the optical bistability in a nanoantenna array is investigated theoretically. The optical response of the array is modeled using a surface integral equation method developed for periodic structures, and the description of the Kerr effect is based on an analytical model. Different behaviors are observed when the distance between the nanoantenna array and the silver layer is changed. Indeed, a modification of the nanoantennas radiative properties permit to control important parameters of the nonlinear response such as the intensity threshold and the area of the hysteresis cycle. The results presented in this article demonstrate that a reflecting surface is a convenient and flexible tool for controlling the operating of nonlinear optical systems based on the Kerr effect.

Keywords Optical bistability · Nanoantenna · Kerr effect · Nonlinear plasmonics · Surface integral equation method

Introduction

Metallic nanostructures support surface plasmon resonances corresponding to the collective excitations of the conduction electrons and exhibit unique optical properties [1]. The coupling between several substructures is a convenient way to control surface plasmon resonance properties [2–4]. In particular, bringing two nanostructures in close proximity results in an enhancement by several orders of magnitude for the electric field in the nanogap, producing a so-called hot spot. This geometry is called a nanoantenna and represents the optical analogous of microwave and radiowave antennas [5–7]. The electromagnetic properties of nanoantennas can be tuned by modifying their geometric parameters (length, shape, and gap dimension) and tailored for specific applications [8]. For instance, optical antennas have been designed for studying quantum systems at the single emitter level [9–11], surface-enhanced Raman scattering [12, 13], and trapping [14–16]. It was shown that the radiative properties of nanoantennas can be controlled using a reflecting metallic surface [17, 18]. For example, this feature has been previously used to enhance the fluorescence and surface-enhanced Raman scattering for molecules close to metallic nanoantennas [19, 20].

Optical antennas are also promising for the observation of nonlinear optical effects which require the high electric field observed in the hot spots [21]. Several studies have discussed multi-photon-excited luminescence [22], second harmonic generation [23–26], third harmonic generation [27, 28], and four-wave mixing [29, 30]. The nonlinear Kerr effect in plasmonic nanostructures coupled to nonlinear materials has also been investigated in several publications [31–35]. Including a Kerr material in plasmonic nanostructures is a convenient way to achieve the control of light by light at the nanoscale and optical memories, transistors, and switches can be designed using this approach [36, 37].

In this article, a new approach to control the Kerr effect down to the nanoscale is introduced. A reflecting surface is used to modify the coupling between the incident light and a nanoantenna array, and the influence on its nonlinear optical response is investigated. The optical response of the array is modeled using a surface integral equation (SIE) method involving a periodic Green's function. The Kerr effect is described using an analytical model which was previously introduced for isolated nanoantennas loaded with a nonlinear material [33] and modified here for the description of nanoantenna arrays close to a metallic surface. In particular, a silver layer is added below the dielectric layer, and the influence of

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the distance between the metallic surface and the nanoantenna array on the nonlinear response is studied in details. The results presented in this article demonstrate that a reflecting surface is a convenient way of controlling the operation of nonlinear optical systems.

Results and Discussion

The article is organized as follows. First, the linear optical response of a nanoantenna array on a SiO_2 substrate is evaluated using a SIE method [38, 39]. The influence of a reflective silver surface on this response is then discussed in details. Finally, the nonlinear response resulting from the Kerr material loaded in the nanoantennas gaps is investigated using an analytical model adapted from [33].

Linear Response of Nanoantennas Array

Let us first consider the linear optical response of a nanoantenna array. The nanoantenna arms are made of silver, and the nanogaps are completely filled with polystyrene (linear refractive index n_0=1.60). The length, width, and height of the rectangular arms are 60, 20, and 20 nm, respectively. The arms are separated by a 10-nm nanogap. The period of the two-dimensional array is 500 nm in both directions. All these parameters are kept constant in the following discussion. The nanoantenna array is deposited on a semi-infinite SiO_2 substrate (refractive index n=1.55). The linear electromagnetic response of this system is computed with the SIE method using a periodic Green's function [38, 39]. This method requires the discretization of the structure in the unit cell only and was found to be very accurate even in resonant conditions [38, 39]. The surface of the plasmonic nanoantenna is discretized with a triangular mesh with a typical side of 5 nm, and the surface of the substrate is discretized with a triangular mesh with a typical side of 25 nm. Indeed, a finer mesh is required to reproduce the significant electromagnetic field variation close to plasmonic nanostructures and nanogaps [40]. The array is excited by a plane wave polarized along the nanoantenna arms and propagating along the normal to the SiO_2 surface. The reflectance of the nanoantenna array is shown as a function of the incident wavelength in Fig. 1b. The dielectric constant of silver is taken from experimental data [41]. A strong increase of the reflectance is observed as the incident wavelength is tuned close to 710 nm, corresponding to the resonant wavelength of the surface plasmon mode supported by the nanoantennas. In the next section, a reflecting silver surface is placed below a finite thickness SiO_2 substrate, and its influence on the optical response of the nanoantenna array is investigated.

Influence of the Reflecting Surface

In order to understand the role of a reflective surface, we first consider a SiO_2 layer without nanoantenna array deposited on a semi-infinite silver substrate (Fig. 2a). When the incident and the reflected waves interfered constructively at the molecule position, the optical intensity is increased. This condition can be satisfied by modifying the dielectric spacer, i.e., the SiO_2 layer thickness. Considering the reflection at the interface of a perfect electric conductor, the optimal thickness corresponding to constructive interference is given by [18]:

$$2d\,n = \left(m + \frac{1}{2}\right)\lambda, \qquad (1)$$

where n is the refractive index of the dielectric spacer (SiO_2 in the present case), d is the dielectric spacer thickness, λ is the incident wavelength in vacuum, and m is an integer. Considering an incident wavelength corresponding to surface plasmon resonance supported by the nanoantenna array discussed previously (λ=710 nm), the first thickness leading to constructive interference is found to be d=114 nm. To confirm this, the electric field intensity is evaluated 10 nm above the SiO_2 layer as a function of its thickness using the SIE method. An intensity maximum is observed for a dielectric layer thickness between 80 and 100 nm. This thickness is slightly shorter than the value predicted by the Eq. (1) since the incoming light penetrates into the silver slab, which is not a perfect conductor at this wavelength, resulting in an additional phase shift [19].

We now include the nanoantenna array on the dielectric substrate and investigate the influence of the silver surface on its optical response. The system is then composed of a nanoantenna array, a SiO_2 dielectric spacer, and a semi-infinite silver slab (Fig. 3a). The reflectance is computed for thicknesses d between 100 and 200 nm. The result is shown as a function of the incident wavelength in Fig. 3b. The different reflectance spectra were fitted with the function:

$$R = 1 - \frac{2A_0}{\pi}\frac{w}{4(\lambda-\lambda_c)^2 + w^2}, \qquad (2)$$

where λ is the incident wavelength, λ_c is the dip wavelength, w is the full width at half minimum, and A_0 is a fitting parameter, with units of a length, related to the dip magnitude. The fits are shown as continuous lines in Fig. 3b. It is found that the reflectance dramatically depends on the dielectric spacer thickness. For the largest studied thickness (d=200 nm), the reflectance does not depend on the incident wavelength and is close to unity. This behavior is easily understood from Fig. 2b, which indicates that the intensity vanishes at the nanoantenna position in this configuration. As a consequence, the surface plasmon resonance supported by the nanoantennas is not excited and does not modify the optical response of the system, leading to a

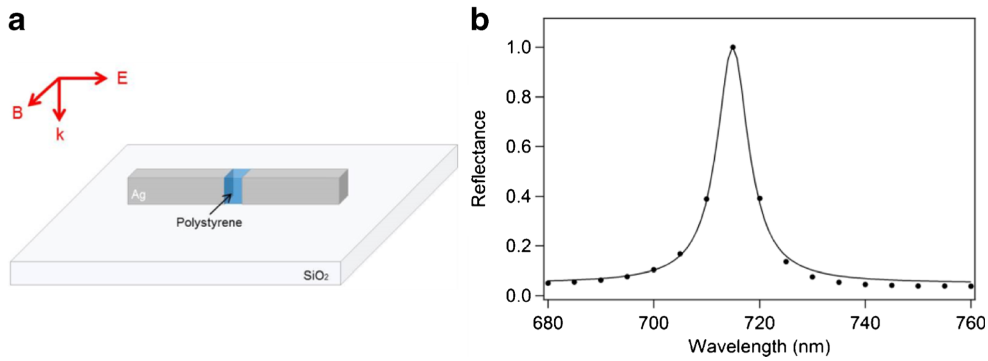

Fig. 1 **a** Schematic of a silver dipolar nanoantenna loaded with polystyrene and deposited on a SiO_2 substrate. **b** Reflectance of a periodic array of silver nanoantennas loaded with polystyrene on a SiO_2 substrate as a function of the incident wavelength. The spatial period is 500 nm. The incident plane wave is polarized along the nanoantenna arms and its wave vector is directed along the normal to the SiO_2 surface. The incident wave is incident from the air side. The continuous line is a fit with a Lorentzian function

flat response for d=200 nm in Fig. 3b. This behavior has already been discussed in the case of a single plasmonic nanorod inside a microcavity [42]. According to Fig. 2b, the coupling between the electromagnetic wave and the periodic array of silver nanoantennas must increase when the thickness d decreases. In this case, the interference between the incoming and the reflected waves are not totally destructive at the nanoantenna position. The excitation of the surface plasmon resonance supported by the nanoantennas results in the formation of a dip in the reflectance spectra (Fig. 3b). As the coupling increases, i.e., as the distance decreases, this dip becomes deeper and broader, revealing that the radiative behavior of the system is indeed modified. The maximum reflectance dip corresponds to the maximum field enhancement in the nanoantenna gaps, as reported in [18]. We will show that this modification of the intensity enhancement in the nanoantenna gap can be used to control the nonlinear response of the nanoantenna array.

Nonlinear Kerr Effect

In this section, the nonlinear Kerr effect in the polystyrene part loaded in the nanoantenna gaps is taken into account. Reference [33] introduces an analytical model to describe the Kerr effect in isolated plasmonic nanoantennas. This model assumes that the electric field distribution is uniform in the nanogap, resulting in a uniform refractive index change. Fig 3a indicates that this approximation is also valid in the present case, where the intensity in the nanogap does not vary by more than a third of the maximum intensity in the nanogap center (Fig. 3a). This previous analytical model is now modified to describe the case of a nanoantenna array close to a reflecting surface with the aim to determine the relation between the incident intensity and the reflectance. To describe the Kerr nonlinearity, the refractive index of the polystyrene is now a function of the intensity [43]:

$$n = n_0 + n_2 I, \tag{3}$$

where n_0 is the linear part of the refractive index, n_2 is the nonlinear Kerr coefficient, and I is the intensity in the Kerr material, i.e., in the nanogap. The intensity I is the intensity of the electric field and I=1/2ε_0|E|^2. The nonlinear Kerr coefficient of polystyrene is n_2=1.14×10^{-12} cm^2/W [44]. This coefficient is positive and the Kerr effect leads to an increase of the refractive index of polystyrene. In order to determine the

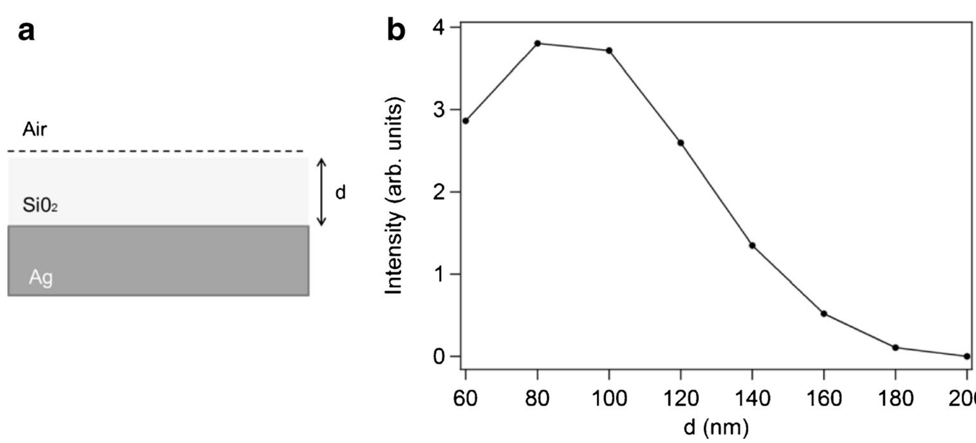

Fig. 2 **a** Schematic of a SiO_2 film on a silver substrate. **b** Intensity as a function of SiO_2 thickness d evaluated 10 nm above the SiO_2 substrate (in air). The wavelength of the incident plane wave is λ= 710 nm

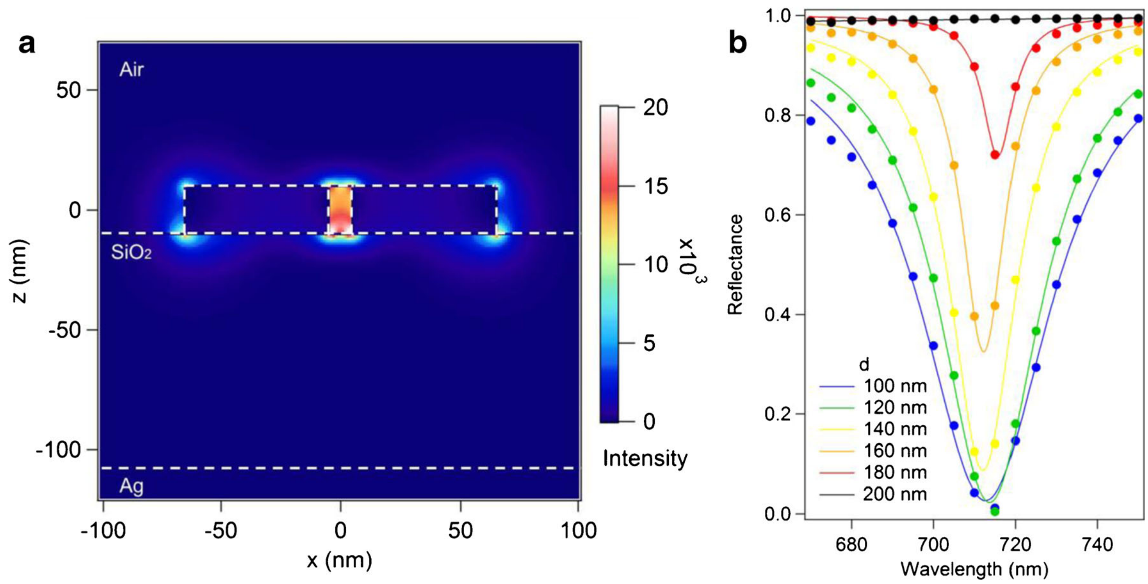

Fig. 3 **a** Near-field distribution of the intensity close to a silver nanoantenna loaded with polystyrene. The nanoantenna is part of an array with a period of 500 nm. The incident wavelength is λ=710 nm. **b** Reflectance for a periodic array of silver nanoantennas loaded with polystyrene on a SiO_2–Ag layered substrate as a function of the incident wavelength for different values of the SiO_2 layer thickness *d*. The continuous lines are fits using Eq. (2)

influence of an increase of the load refractive index on the optical response of the nanoparticle array, computations have been performed considering different values of this refractive index for two different SiO_2 layer thicknesses, namely d=100 and 180 nm. Figure 4 shows the intensity enhancement factor I/I_0, where I_0 is the incident intensity and I is the intensity at the nanogap center (Fig. 3a), and the reflectance spectra for different values of the refractive index of the material loaded in the gap. It is clearly observed that an increase of the refractive index leads to a redshift of the surface plasmon resonance supported by the nanoantennas.

The resonance wavelength λ_I can be expressed as [33]:

$$\lambda_I = \lambda_c + \alpha n_I, \tag{4}$$

where λ_c is the dip wavelength computed considering only the linear refractive index of the polystyrene, n_I is the variation of the refractive index, and α is a weighting coefficient, which can be extracted using a fitting procedure. The nonlinear Kerr effect in the polystyrene load is expected to have a similar detuning effect and to also modify the resonant wavelength of the nanoantenna array. Taking into account this effect, Eq. (4) can be rewritten as:

$$\lambda_I = \lambda_c + \alpha n_2 I. \tag{5}$$

The data reported in Fig. 4 allow determining the relation between the reflectance and the enhancement factor for a given configuration at different wavelengths; a linear relation is observed between the enhancement factor and the reflectance. This allows us to deduct a relation between the reflected intensity I_{ref} and the intensity in the gap:

$$I = \beta I_{\mathrm{ref}} + \gamma. \tag{6}$$

Finally, the reflected intensity becomes:

$$I_{\mathrm{ref}} = I_0 \frac{4(\lambda - \lambda_I)^2 + w^2 - \frac{2A_0 w}{\pi}}{4(\lambda - \lambda_I)^2 + w^2}, \tag{7}$$

with $\lambda_I = \lambda_c + \alpha n_2(\beta I_{\mathrm{ref}} + \gamma)$. Equation (7) is a third order algebraic equation that provides the reflectance as a function of the incident intensity. All the parameters involved in this relation are functions of the dielectric spacer thickness *d*. In the following, we use the relation (7) to determine the influence of the reflecting silver surface and the SiO_2 layer on the nonlinear response of the nanoantenna array.

Figure 5 shows the reflectance as a function of the incident intensity for different incident wavelengths and SiO_2 layer thicknesses *d*. The first considered wavelength is λ=710 nm corresponding to the resonant wavelength of the surface plasmon mode supported by the nanoantennas (see Fig. 1). For the two considered thicknesses, a modulation of the reflectance is observed, but the system does not support optical bistability (Fig. 5a). Indeed, it was demonstrated in the case of an isolated nanoantenna that optical bistability can be observed only if the relation

$$\lambda > \lambda_c + \frac{\sqrt{3}w}{2} \tag{8}$$

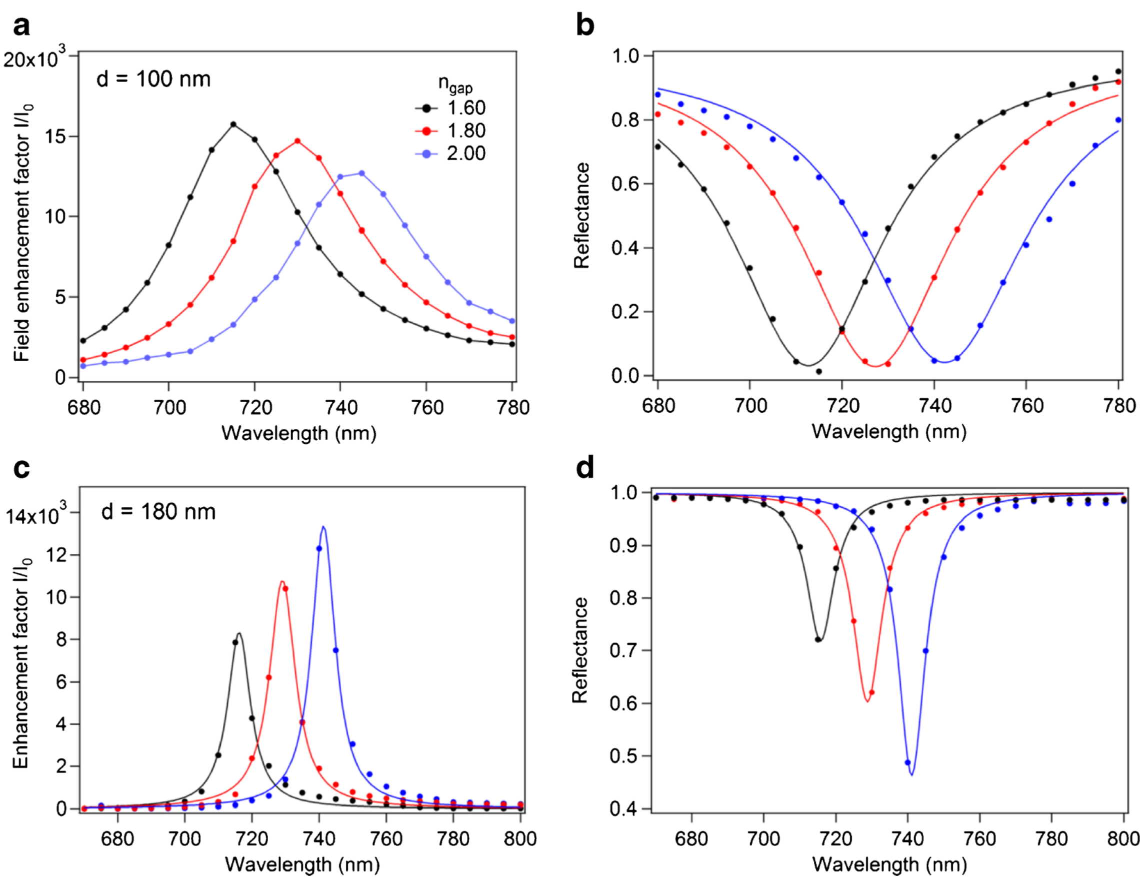

Fig. 4 **a**, **c** Enhancement factor I/I_0 evaluated at the nanoantenna gap center and **b**, **d** reflectance computed for different values of the optical refractive index of the loaded material n as a function of the illumination wavelength. Two SiO2 layered thicknesses are considered: **a**, **b** d=100 nm or **c**, **d** d=180 nm

is satisfied [33]. This is clearly not the case here. Eq. (8) indicates that a longer wavelength is required for the observation of optical bistability. Figure 5b shows the reflectance as a function of the incident intensity for an incident wavelength (λ=730 nm) slightly longer than the linear resonant wavelength. Even though only a modulation of the reflectance is observed for d=100 nm, optical bistability is clearly visible for d=180 nm. Note that the unstable branch is not shown in Fig. 5. The different behaviors are explained by the modification of the resonance width. The presence of the reflecting surface modifies the radiative properties of the nanoantenna array and the quality factor of the surface plasmon resonance (see Fig. 4) [18]. The resonance is narrower for d=180 nm, and optical bistability is observed in agreement with Eq. (8). Finally, an even longer wavelength is considered (λ=770 nm). Figure 5c shows that at this wavelength, optical bistability is observed for the two considered

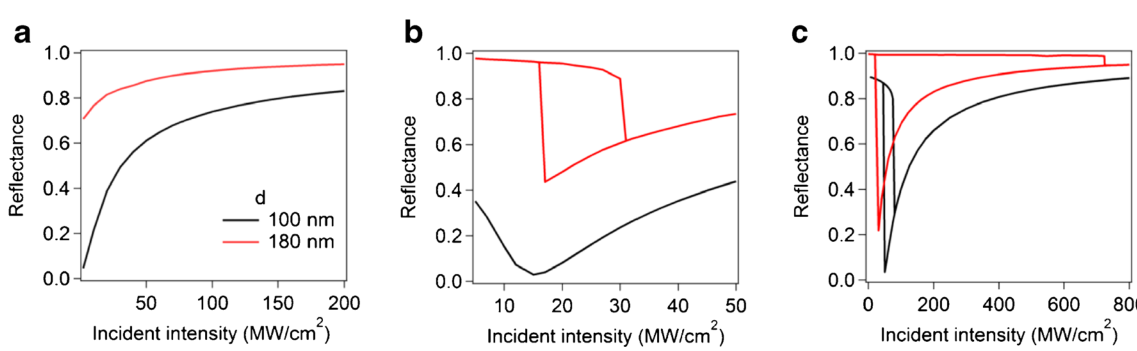

Fig. 5 Reflectance as a function of the incident intensity evaluated for a SiO2 thickness d=100 nm (*black curves*) and d=180 nm (*red curves*) considering the influence of the polystyrene Kerr effect on the nanoantenna array response, as discussed in the text. The incident wavelength is **a** λ=710 nm, **b** λ=730 and λ=770 nm

thicknesses *d*. Interestingly, important parameters such as the intensity threshold and the area of the hysteresis cycle depend on the distance between the nanoantenna array and the reflecting surface. Despite the simplicity of the proposed approach, a clear modification of the intensity threshold is observed. Indeed, Fig. 5c shows that a small variation (80 nm) of the distance between the silver film and the nanoantenna array modifies the intensity threshold by ~10. Note that the intensity threshold depends on both the resonance width *w* and the field enhancement in the nanogap (see Fig. 4). Furthermore, it is worth noting that optical bistability can be achieved at lower incident intensities than those reported for isolated nanoantennas [33]. These results clearly demonstrate that a reflecting surface can be used to control the nonlinear response of plasmonic nanoantenna array. Nanoantenna arrays loaded with nonlinear materials were proposed as optical memories, transistors, and switches at the nanoscale [36, 37]. The results presented in this article show that a reflecting surface is a convenient tool for tailoring their optical properties and controlling their operation. From an experimental point of view, the addition of a polystyrene load in the nanoantenna gaps can be very challenging. One possibility is then to completely cover the nanoantenna array with a polystyrene layer. In this configuration, the Kerr effect is significant only in the nanoantenna gap, where the electromagnetic is enhanced by several orders of magnitude.

Conclusions

In summary, the influence of a reflecting surface on the nonlinear response of a nanoantenna array was investigated using a surface integral equation method and an analytical model for describing the Kerr effect in nanoantennas loaded with a nonlinear material. The influence of a silver layer on the nonlinear response of the nanoantenna array was investigated in details. Interestingly, it was shown that different behaviors are observed when the distance between the nanoantenna array and the silver layer is modified. This feature can be used to control the nonlinear response of metamaterials combining plasmonic and Kerr materials. The strength of the proposed approach is its simplicity and the possibility to extend it to any plasmonic system, such as the nonlinear nanoshells (nonlinear nanocores covered with silver shells) introduced by Argyropoulos et al. [37] or the hybrid metal-dielectric nanoantennas (which are composed of crystalline silicon and silver nanoparticles) proposed by Noskov et al. [36] for example. Furthermore, the method proposed in this article can be combined with other ones in order to achieve efficient optical nanomemories or nanoswitches. For example, it was reported that including a gain medium, which can be supplied by molecules or quantum dots, boosts the performance of plasmonic devices [45–47].

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