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Enhanced optical bistability with film-coupled plasmonic nanocubes

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Colloidally synthesized nanocubes strongly coupled to conducting films hold great promise for enhancing different nonlinear optical processes. They exhibit a robust and sensitive scattering response that can be easily controlled by their geometrical and material parameters. Strong local field enhancement is generated at the gap regions between the nanocubes and the metallic film. We show that strong optical bistability and all-optical switching behavior can be obtained by loading these nanogaps with Kerr nonlinear materials. Relatively low input intensities are required to obtain these nonlinear effects. The proposed design can lead to efficient, low-power, and ultrafast all-optical memories and scattering nanoswitches. © *2014 AIP Publishing LLC*.
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Integrated photonic devices that possess a bistable response are the key elements for all-optical computing applications. The weak nature of the nonlinear optical interactions places a huge limit to the practical implementation of optical bistability; extremely high pump intensities are in fact required to obtain noticeable effects.[1] Although it is possible to relax the requirement on the input intensity by using very high quality factor cavities, i.e., extended Fabry–Pérot gratings and photonic-crystal resonators,[2–6] this is extremely detrimental to the operational bandwidth of the device and its integration to nanoscale dimensions.

Recently, significant research efforts have been devoted to control and enhance light-matter interactions at subwavelength scales by means of plasmonic platforms[7,8] In particular, extreme coupling between nanoparticles or nanoparticles and conducting films has shown to increase local field intensities up to three orders of magnitude with respect to the incident radiation.[8] In addition, the development of bottom-up fabrication techniques has granted simple and cheap access to nanometric-gap structures.[9,10] Colloidally synthesized metal nanoparticles can, for example, be adsorbed on large area self-assembly organic layers only few nanometers thick, deposited on conducting substrates.[11] The resonance of such systems is strongly dependent on the dielectric thickness and requires very small separation distances in order to span the entire visible spectrum. By using colloidal silver nanocubes, it is possible to introduce a better control on the resonances and even push them down to the near-infrared.

The efficient local field enhancement and flexible tunability serve as an excellent platform to boost several nonlinear optical processes,[12] which can make nonlinear optical effects accessible with much lower input intensities and within nanosized structures, in contrast to the previous designs. Different nonlinear materials may be included within the plasmonic "hot-spot" field regions, leading to effectively enhanced nonlinear optical processes, such as second harmonic generation ($\chi^{(2)}$ media)[13–18] and optical bistability ($\chi^{(3)}$ media).[19–26]

Although the intrinsic absorption of metals is considered to be detrimental for most linear applications, it is generally far less critical for nonlinear applications, where conversion rates are expected to be a few percent. Moreover, the role of metals in nonlinear optics applications goes beyond their ability to support surface plasmon modes that allow the light to become strongly confined in deeply subwavelength volumes. Metals possess third-order nonlinear susceptibilities that are orders of magnitude larger than dielectric materials.[1,27] As a result, a major research effort has been targeted toward metallo-dielectric composites[28,29] and metamaterial composites,[30–32] including structured films and surfaces.[33–36] However, bulk metallic structures are characterized by high reflectivity and the fields cannot efficiently couple inside them, leading to poor field enhancement and, as a result, weak nonlinear optical effects.

Although, moderate threshold input pump intensities were still required in all the previous nonlinear plasmonic works, nevertheless, it is expected to achieve even more pronounced low-power nonlinear optical effects, in case the plasmonic modes can be strongly confined and coupled in extremely subwavelength regions loaded with nonlinear materials, such as narrow waveguides, ultrasmall nanocavities, and nanogaps. This will lead to an even better compactness of nonlinear optical designs, bringing the highly anticipated ultrafast and low-power integrated nanophotonic logical circuits one step closer to their practical realization.

In this Letter, we will focus our study to film-coupled nanocubes[11,37–39] loaded with nonlinear materials to achieve all the aforementioned goals. It will be demonstrated that this particular plasmonic design constitutes an ideal platform towards the experimental realization of compact, ultrafast, and low-power nonlinear plasmonic systems with instantaneous nonlinear optical responses. Furthermore, the bottom-up chemical synthesis approach is ideal for large-scale practical implementation of such structures. The nanocubes are single crystalline and can be coupled to atomically smooth epitaxially grown silver films[40] or ultrasmooth template-stripped

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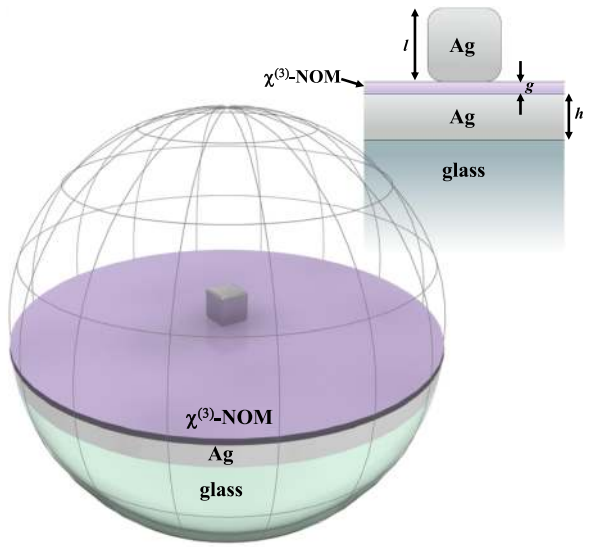

FIG. 1. Schematic of the film-coupled nanocube structure. The substrate is composed of glass. Kerr nonlinear material is loaded at the spacer layer, which separates the nanocube with the silver film. The dimensions of the nonlinear system are: $l = 80\,\text{nm}$, $h = 50\,\text{nm}$, and $g = 2\,\text{nm}$.

metallic films.[41] This is expected to drastically reduce the effective damping constant of the metallic film and enables experimental results which are in close agreement with the numerical simulations. This property makes the proposed plasmonic system even more appealing to enhance different active and nonlinear optical processes, where the lasing or nonlinear processes are much more sensitive to losses. In this work, we will consider Kerr $\chi^{(3)}$ nonlinear materials embedded into the spacing layer between the nanocube and the metallic film. The nonlinear analysis will be limited to third-order Kerr nonlinear effects, such as optical hysteresis and all-optical switching, but the structure is expected to be advantageous in several other nonlinear applications, i.e., second/third-harmonic generation and four-wave mixing.

The proposed plasmonic design is shown in Fig. 1, where a silver nanocube is positioned above a dielectric-coated metallic (Ag) film. The plasmonic nanocube is strongly coupled to its electromagnetic mirror image, effectively leading to the formation of a nanodimer with a deeply subwavelength gap in which a nanocavity is formed characterized by an exceptionally strong field enhancement. The ultrathin dielectric spacing layer is constituted by a Kerr nonlinear optical material ($\chi^{(3)} - NOM$) with relative permittivity $\varepsilon = \varepsilon_L + \chi^{(3)}|E|^2$, with $\varepsilon_L = 2.2$, and $\chi^{(3)} = 4.4 \times 10^{-18}\text{m}^2/\text{V}^2$, which is typical nonlinear material values of semiconductors and polymers;[1] $|E|$ is assumed to be the magnitude of the mean value of the local complex electric field inside the nanogap. Note that the corners of the nanocubes are slightly rounded in order to avoid numerical artifacts and to be in agreement with a potential experimental realization of such plasmonic system.[11,39] Silver losses are fully taken into account in our analysis by using a silver dielectric function obtained from experimental data.[42] The Ag film is placed over a semi-infinite glass substrate with refractive index $n = 1.47$, and the dimensions of the plasmonic system are chosen (with respect to the inset of Fig. 1) to be: $l = 80\,\text{nm}$, $h = 50\,\text{nm}$, and $g = 2\,\text{nm}$. Although the gap is highly subwavelength, the analysis can still be performed without taking into account nonlocal[38,43–46] or quantum[47–49] effects which have been shown to severely affect the scattering response and limit the field enhancement in similar plasmonic structures with only a few Angstrom thickness (subnanometer) gaps (typically $g < 0.5\,\text{nm}$).

The three-dimensional (3D) film-coupled plasmonic nanocube structure of Fig. 1 is simulated using the finite element method (COMSOL Multiphysics). In this Letter, the device is always excited by transverse-magnetic (TM) polarized plane waves impinging at an incident angle of $\theta = 75^{\circ}$ with respect to normal. However, the scattering response is fairly independent of the angle of incidence and polarization of the plane wave excitation, as it was demonstrated earlier in Refs. 11 and 39. First, we consider the linear operation of the plasmonic nanocube system, where a conventional dielectric is loaded at the spacer layer with relative permittivity $\varepsilon_L = 2.2$. We numerically compute the scattering cross-section (SCS) of this structure and normalize its value to the dimensions of the nanocube, in order to compute the SCS efficiency of the entire plasmonic system. The result is presented in Fig. 2, where a sharp narrowband scattering response is obtained with a pronounced scattering peak around $\lambda \simeq 1.07\,\mu\text{m}$. The quality factor of this resonant response is approximately $Q \simeq 80$, typical value for plasmonic resonances. Standing waves are rapidly built inside the nanogap at this frequency point and the computed electric field enhancement distribution can be seen in the inset of Fig. 2. The cavity plasmonic mode is caused by the built-up of the standing waves, which directly leads to enhanced localized electric fields inside the nanogap with a maximum enhancement factor of about 200, as it is clearly demonstrated in the inset of Fig. 2. The rapid increase in scattering efficiency can also be translated into an increase in absorption strength, leading to efficient plasmonic absorber designs.[11]

Hence, we will employ the large local field enhancement and high SCS efficiency to boost nonlinear optical processes. To this end, Kerr nonlinear $\chi^{(3)}$ material is loaded at the spacer layer between the nanocube and the film with parameters and geometry described before. In general, the intensity

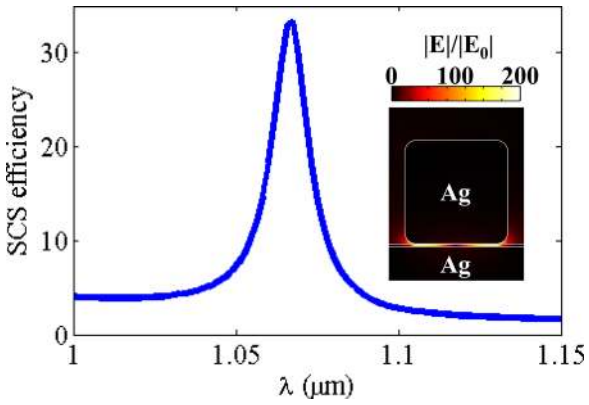

FIG. 2. SCS efficiency of the linear nanocube plasmonic system varying with the input radiation's wavelength. A dielectric material is loaded at the spacer layer with relative permittivity $\varepsilon_L = 2.2$. The strong field enhancement is depicted in the inset of this figure and is mainly localized in the spacer layer.

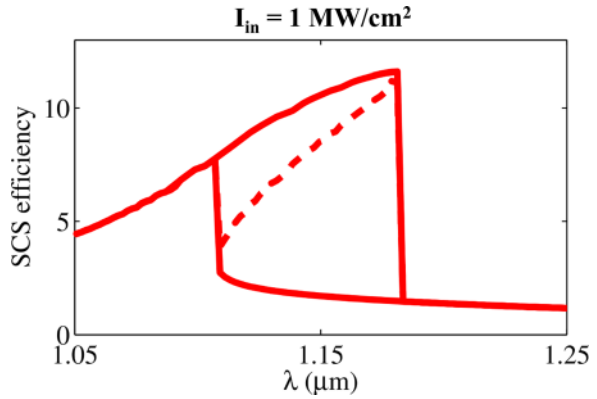

FIG. 3. Strong bistability in the scattering performance of the nonlinear plasmonic nanocube system with geometry shown in Fig. 1. The SCS efficiency is plotted versus the wavelength and the device is pumped with low input intensity $I_{in} = 1\text{MW/cm}^2$. The unstable branch of the bistable curve is shown with a dashed line.

dependent permittivity of Kerr nonlinear materials can lead to optical hysteresis and all-optical switching responses.[1] Here, we apply a mean-field approximation to monitor the fields inside the nanocavity and we compute the SCS efficiency of this nonlinear plasmonic system for different applied input intensities. The graphical solution approach is used to post-process all these results and calculate the solutions of the nonlinear problem.[1] The bistable SCS efficiency performance of the nonlinear structure under study is computed and plotted in Fig. 3 versus the wavelength of the input radiation for relatively low input pump intensity $I_{in} = 1\text{MW/cm}^2$. Strong hysteresis in the scattering efficiency is obtained and the nonlinear scattering spectrum is spread in a relative broad frequency range compared to the linear results presented in Fig. 2. In the same plot, we also show the calculated unstable branch of the predicted nonlinear response, plotted as the dashed line. This is another mathematical solution of the nonlinear problem, but it is not reachable from the two stable branches (solid lines in Fig. 3). An exceptionally strong bistable performance is obtained in the nonlinear SCS efficiency spectrum, which can become even wider if we increase the input pump intensity impinging at the plasmonic nanocube. The input intensity required in our case is several orders of magnitude lower compared to nanoantennas embedded or placed over Kerr nonlinear materials,[22–26,50] where bistability and modulation were predicted in the scattering or reflection responses with input intensities with approximate values in the range of $I_{in} \simeq 0.5 - 1\text{GW/cm}^2$. The key distinction of the presented work compared with the previously published nonlinear plasmonic systems is that in our plasmonic configuration we can precisely place the nonlinear material in the nanogap region, where the highest fields are obtained.

Next, we fix the wavelength of the nonlinear operation to $\lambda = 1.105\,\mu$m, slightly above the resonant scattering peak of the linear operation (Fig. 2). We monitor the scattering response of the nonlinear plasmonic system as the input radiation intensity is varied. The device always has the same dimensions and parameters, presented earlier in Fig. 1. Strong low-power all-optical scattering switching behavior is obtained, when the input power is increased or decreased, and the response is plotted in Fig. 4. Note that in case the gap thickness of the proposed structure is increased, the field enhancement will be decreased inside the formed nanocavity.[11,39] However, we have calculated that still low input intensities $I_{in} \simeq 3 - 6\text{MW/cm}^2$ are needed to excite bistable performance for the same nonlinear plasmonic system but with increased gap thickness $g = 5\,\text{nm}$.

The reported broad hysteresis loop demonstrates an efficient way to design a compact low-power optical nanoswitch with high sensitivity to relative low input pump intensities. When we increase the input intensity of the impinging radiation, the SCS efficiency remains almost constant, taking particularly low values. However, when the threshold input intensity value $I_{up} = 820\text{kW/cm}^2$ is reached, the SCS efficiency experiences an abrupt "jump" and the device now operates at the upper branch of the scattering hysteresis curve. If we start to decrease the input intensity, after the threshold intensity value has been reached, the nonlinear structure will keep its high SCS efficiency operation, which will be even further increased in this particular plasmonic configuration. The SCS efficiency will always be higher than 10 in the upper scattering branch, which is approximately four times higher compared to the SCS efficiency of the lower scattering branch of the hysteresis curve. Only when we reach low input pump intensities with values less than $I_{down} = 180\text{kW/cm}^2$, the SCS response will return to its initial low value, similar to the performance in the beginning, when we just started increasing the input intensity. Indeed, this nonlinear system can exhibit memory and it can operate as an efficient nanoswitch with the OFF-mode operation located at the lower scattering branch (increasing pump intensity) and the ON-mode operation positioned at the high scattering branch (decreasing pump intensity). If illuminated with a continuous wave laser, the plasmonic optical switch can close or open depending on the intensity of the input radiation,[1,20] similar to the well-established logic circuits in semiconductor electronic components.

We envision that the integration of these compact nonlinear plasmonic elements with other planar plasmonic

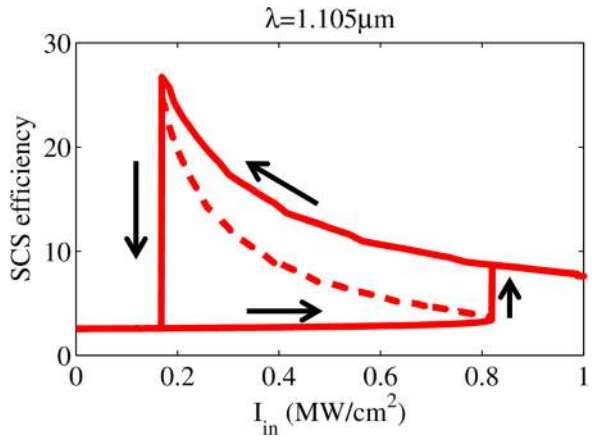

FIG. 4. Bistable SCS efficiency as a function of the input pump intensity of the nonlinear nanocube structure shown in Fig. 1 and monitored at $\lambda = 1.105\,\mu$m. The unstable branch of the curve is shown with the dashed line. Strong scattering hysteresis is obtained with the proposed nonlinear plasmonic system, leading to efficient all-optical scattering switching behavior.

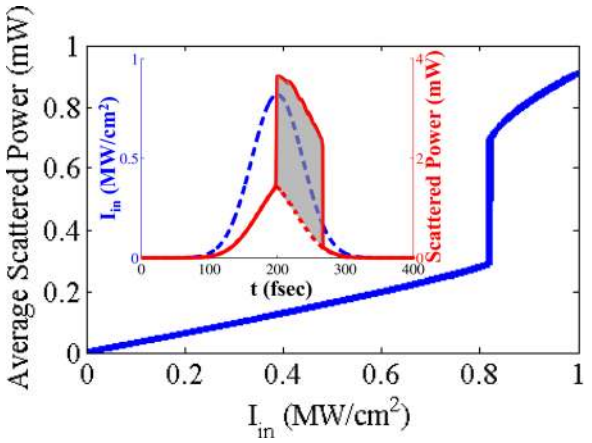

FIG. 5. Average scattered power of the nonlinear plasmonic device when the input intensity of the radiation is increased. The femtosecond Gaussian pulse (blue dashed line) used for the pulsed laser and the calculated instantaneous scattered power for $I_{in} = 819\text{kW/cm}^2$ (red dashed line) and $I_{in} = 820\text{kW/cm}^2$ (red solid line) are plotted in the inset. A pronounced scattering "jump" is observed for both cases when the input intensity of the pumped pulse acquires higher than the threshold values.

metasurfaces will lead to complex all-optical integrated nanophotonic systems with ultrafast, broadband, and low-power functionalities. The simplicity involved in the fabrication of the proposed devices, mainly based on straightforward colloidal self-assembly processes, may pave the way to a potential experimental verification of the proposed nonlinear plasmonic system. We theoretically predict the scattering response of the proposed nonlinear plasmonic device based on a simple experimental set-up and the results are depicted in Fig. 5. A femtosecond pulsed laser with a repetition rate of 12.5 ns, a full-width at half-maximum (FWHM) of 150 fs, and a peak intensity of $I_{in} = I_{up} = 820\text{kW/cm}^2$ is launched at the proposed structure and its intensity response is plotted in the inset of Fig. 5 (blue dashed line). The predicted instantaneous scattered power of the nonlinear device for two laser excitations with slightly different peak intensities of $I_{in} = 819\text{kW/cm}^2$, below the threshold, and $I_{in} = 820\text{kW/cm}^2$, above the threshold, are also shown in the inset of Fig. 5 as the red dashed and solid lines, respectively. It is interesting that with just a small change in the input intensity of the excitation pulse, the instantaneous scattered power experiences an abrupt increase, which is highlighted by the shadowed region in the inset of Fig. 5.

It is not possible to measure the instantaneous SCS in an experiment; however, we can easily monitor the average scattered power as we change the input intensity of the pulsed laser with the specifications presented before. The result is demonstrated in Fig. 5, where a sharp "snap" in the average scattered power is observed when the input intensity of the pumped pulse acquires higher than the threshold values, as it was predicted before in Fig. 4. The femtosecond pulsed laser used to obtain these results has a very low peak intensity value $I_{in} = 1\text{MW/cm}^2$, which can be easily increased by several orders of magnitude with the existing high-energy pulsed lasers. As a result, the higher is the intensity peak of the input laser, the sharper and stronger will be the "jump" in the average scattered power and, equivalently, the broader will be the optical scattering hysteresis response of the designed nonlinear plasmonic system.

All the aforementioned nonlinear effects require very low input intensities to be triggered compared to the previous examples in the literature of nonlinear plasmonic devices. The proposed device can lead to low-power ultrafast nonlinear components which are expected to be revolutionary elements in the future integrated nanophotonic circuits. The realistic ohmic losses of the presented nonlinear plasmonic system have been taken into full account in our calculations. Therefore, we do not foresee any difficulty in experimentally realizing the demonstrated nonlinear effects in the near future and we suggest a simple experimental set-up to observe the predicted strong nonlinear processes. Furthermore, the scattering response of the proposed device may be tuned to a large degree by just altering the size and shape of the structure,[11] providing additional flexibility in the design of the proposed nonlinear device. Note that the field enhancement will be decreased by approximately 10 times in case the metallic film is removed,[50] which will lead to the disappearance of the key property proposed in the current work, i.e., strong film coupling. As a result, 100 times higher input intensities will be needed to trigger nonlinear effects, such as bistability, in this alternative nonlinear plasmonic structure.

The proposed nonlinear plasmonic system can still exhibit bistable performance, even if the dimensions of the structure are relaxed or different dielectric and metallic materials are used. For example, we may use different nonlinear materials in the spacer layer. Oxides can be deposited with atomic layer deposition (ALD) methods.[51] Polymer films can be prepared to have very small thicknesses with a layer-by-layer deposition process.[11,39,52] In general, polymers can have very high nonlinear properties[53] and they can be ideal materials for the spacer layer in the experimental verification of the proposed concept. Thin chalcogenide films with high nonlinear properties can be prepared with lithography and placed adjacent to metallic parts.[54] Moreover, nanolithography has been widely used to create silicon-based plasmonic structures for on-chip photonic applications,[55,56] which may lead to an efficient alternative design of the proposed structure. Another nonlinear material with a large nonlinear coefficient is high conductivity ITO,[50] which may be placed in the nanogap of our structure. Alternatively, two dimensional ultrathin materials, such as graphene, can be directly placed at the nanogap between the metallic parts.[57] It is interesting that graphene has recently found to possess extremely high nonlinearity at similar frequency ranges with the presented work.[58] To conclude, we believe that the presented results can lead to plasmonic structures with robust self-tunable nonlinear operation and ultrafast, almost instantaneous, scattering switching response. Our findings may have a plethora of potential future practical applications, providing viable ways to produce active integrated nanophotonic components, such as low-power nanomemories and all-optical scattering switches.

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