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# Nonlinear Plasmonic Cloaks to Realize Giant All-Optical Scattering Switching

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Here we extend the reach of Fano resonant coupling by combining this concept with cloaking and plasmonic resonances in a single nonlinear nanoparticle, in order to realize giant all-optical scattering nanoswitches controlled by moderate pumping intensities. We show that a core-shell nonlinear plasmonic particle may be designed to abruptly switch from being completely cloaked to being strongly resonant, with up to a 40 dB cross-sectional difference. Self-tunable optical cloaks and resonant scatterers are envisioned for use as efficient all-optical switches and nanomemories.

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Considerable attention has been recently paid to the concept of Fano scattering resonances, produced by the strong coupling between a bright and a dark nonradiating mode supported by a complex subwavelength plasmonic system [1–5]. These resonances can produce sharp spectral signatures and large, localized field intensities, features of great interest for sensing applications. Fano resonances are usually based on the coupling of different, orthogonal scattering orders, such as the electric (bright) and the magnetic dipole or quadrupole (dark) modes of a subwavelength system, which implies that the Fano signature may be usually observed only over a portion of the angular spectrum, in which the two modes interfere [1–5].

In a rather different context, there has recently been a large interest in manipulating the scattering signature of objects using metamaterials and plasmonics. Low scattering and invisibility may be induced with various cloaking techniques, including transformation-optics [6] and scattering cancellation [7–9] approaches. Conversely, small objects may support bright scattering resonances if coupled to plasmonic materials, whose collective electron excitation may produce total scattering cross sections (SCS) much larger than the physical size of the object [10]. It is interesting that homogeneous plasmonic shells may be tailored to achieve both cloaking [7] and resonant scattering [11], of special interest at optical frequencies at which noble metals naturally support plasmonic features [10–12]. The resonant dipolar scattering state is typically the bright mode involved in plasmonic Fano resonances, in which it is usually coupled to higher-order modes that are less efficient in terms of radiation and scattering. The negative polarizability of a plasmonic shell, however, can be tailored also to suppress the electric dipole radiation from a particle [7], creating an alternative "dark" scattering state. Different from higher-order resonances, plasmonic cloaking is an inherently nonresonant phenomenon [13] and is therefore characterized by a much broader bandwidth [14] compared to dark resonant modes and a much reduced level of stored electromagnetic energy. Its inherent robustness has made possible a recent experimental verification at microwave frequencies [15].

One may wonder whether the dual phenomena of cloaking and resonant scattering based on plasmonic shells may be used as, respectively, the dark and bright modes to induce Fano-like scattering signatures in small particles. One of the advantages of this idea would be that the two scattering states do not need to pertain to different scattering orders and may be both induced within the same electric dipolar scattering mode, observable in the near and far field for any position of the observer. Moreover, it may be easier to induce both scattering states in a subwavelength particle, without relying on higher-order resonances that are inherently very sensitive to losses and disorder due to the larger stored energy associated with them. We may also expect that resonant fields may be induced inside the cloaking layer, combined with very low scattering in the surrounding environment, based on the fact that plasmonic cloaks have been proposed for noninvasive sensing applications [16]. These strongly enhanced fields may be particularly attractive to boost optical nonlinear effects present in the metallic shell or in the core material [17–19]. This exotic scattering response, however, cannot be achieved by applying conventional plasmonic cloaking schemes [13], since its frequency dispersion is too slow to be used in a Fano-like application. We put forward here the concept of combining these exciting ideas and realize a practically viable nonlinear cloak that may provide strong scattering variations controlled by the applied light intensity.

Consider the geometry shown in Fig. 1(a), consisting of a subwavelength plasmonic shell (gray region) with radius $a = 30$ nm surrounding a dielectric nanosphere [yellow (light gray)] with radius $a_c = \eta_c a$ and permittivity $\varepsilon_c = 2.2$. We assume a lossless Drude permittivity dispersion for the plasmonic shell with $\varepsilon_s = \varepsilon_\infty - f_p^2/[f(f + i\gamma)]$, $f_p = 2175$ THz, $\gamma = 0$ THz, and $\varepsilon_\infty = 5$, based on experimentally retrieved silver dispersion [20], in which for now we neglect absorption. The overall SCS of this

 

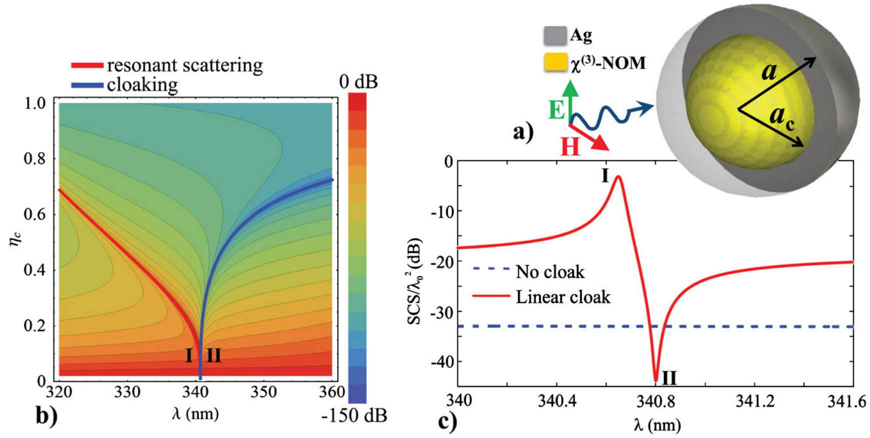


FIG. 1 (color online). (a) A core-shell nonlinear particle illuminated by an impinging beam. (b) Contour plot of the normalized SCS in the linear regime, as a function of wavelength and aspect ratio. In this case, the dielectric core [yellow (light gray) region in (a)] has permittivity $\varepsilon_c = 2.2$. Cloaking [blue (dark gray) line] and resonant scattering [red (gray) line] conditions are also shown. (c) Normalized scattering response versus wavelength (solid red line) for $\eta_c = 0.1$, compared to a bare dielectric sphere with the same size (dashed blue line).

composite particle is shown in Fig. 1(b) as a function of the aspect ratio $\eta_c$ and of the wavelength of operation below the plasma frequency of the metal $f_p$. It can be seen that for large aspect ratios, corresponding to a thin shell, low scattering may be achieved for longer wavelengths with a rather broadband response, significantly separated in frequency from the resonant SCS region, arising for shorter wavelengths. This operation is consistent with our previous works on cloaking (e.g., [7]), in which we aimed at keeping scattering resonances away from the cloaking frequency band to maximize the bandwidth of operation [13].

In the lossless limit considered here, and in the notation of Ref. [11], cloaking arises when $U_1^{\mathrm{TM}} = 0$, which may be written in the long-wavelength regime as the quasistatic condition

$$\eta_c^3(\varepsilon_c - \varepsilon_s)(2\varepsilon_s + 1) + (\varepsilon_c + 2\varepsilon_s)(\varepsilon_s - 1) = 0. \quad (1)$$

Conversely, a resonant peak arises for $V_1^{\mathrm{TM}} = 0$, which provides

$$2\eta_c^3(\varepsilon_c - \varepsilon_s)(\varepsilon_s - 1) + (\varepsilon_c + 2\varepsilon_s)(\varepsilon_s + 2) = 0. \quad (2)$$

The dispersion of these equations is plotted in Fig. 1(b), as the solid red (gray) and blue (dark gray) lines, respectively, which follow the minima and maxima of the calculated SCS, confirming that the electric dipole is mostly contributing to the scattering of the particle in all regions of the plot.

For smaller ratios $\eta_c$ (or thicker shells), the two lines converge to the same condition [points I and II in Fig. 1(b)], and the cloaking bandwidth drastically decreases and merges with the scattering resonance. This is consistent with Eqs. (1) and (2), which for small values of $\eta_c$ interestingly converge to the same condition:

$$\varepsilon_c = -2\varepsilon_s, \quad (3)$$

which represents a quasistatic Fano-like resonance or antiresonance condition for subwavelength core-shell particles. Equation (3) is reminiscent of, but very different from, the usual resonant condition of a homogeneous plasmonic sphere [the limit of Eq. (2) for $\eta_c \rightarrow 0$ or 1]. It refers to an internal resonance supported at the interface between the core and the shell material, arising in the limit in which the core radius is much smaller than the shell. This internal resonance strongly interferes with the plasmonic resonance supported under the same quasistatic condition (3), respectively, leading to very large and very small scattering. In this frequency region, a narrowband Fano-like response is obtained due to the closely spaced interaction of the cloaking dip and the resonant peak, which act as coupled dark and bright scattering states.

Different from conventional Fano plasmonic resonances, for which the "bright" dipolar mode strongly interferes with the "dark" quadrupolar or magnetic mode [1], here the two interfering states are associated with the same scattering mode, the $\mathrm{TM}_1$ spherical harmonic, corresponding to a bonding resonant state (dipolar resonance) and an antibonding nonresonant state (dipolar cloaking). This implies that, even in the case of a single core-shell particle, the Fano signature may be detected at *any angle of observation* and observed also in the *total SCS*. Similar dipole-dipole based Fano-type resonances have also been discussed in Ref. [21] in a different context and for different geometries. The Fano-like scattering response of the proposed structure is shown in Fig. 1(c), where the normalized SCS is shown (solid red line) for $\eta_c = 0.1$, compared to a dielectric particle with same radius $a$ (dashed blue line). In a very narrowband wavelength range, the normalized SCS can go from $3/(2\pi)$ [$\sim -3$ dB, point I in Fig. 1(b)] to almost zero [$\sim -44$ dB, point II]. When considering realistic losses, as we show in Ref. [22], the operational bandwidth of this Fano response expectedly broadens and the scattering excursion is reduced, but the main operation and concept of Fano-like response remain qualitatively unchanged.

In essence, the plasmonic cloaking effect becomes very narrowband when coupled to a plasmonic resonance under condition (3), producing Fano-like antiresonance features. The corresponding field distribution at the two wavelengths of interest is plotted in Fig. 2. In both scenarios the fields around and inside the dielectric core are very similarly confined and largely enhanced at the two wavelengths, but the outside fields are dramatically different: the bright resonant state has a large scattering efficiency [Fig. 2(a)], whereas in the cloaked state the fields are mostly unperturbed, even right around the shell [Fig. 2(b)]. It is interesting that for an observer sitting inside the core region, or near the internal interface, the fields are uniformly boosted all across the wavelength spectrum of interest, without any special sign of antiresonance behavior, but for an observer just around the plasmonic shell the scattering signature can dramatically change from a bright to an invisible state, as a function of the wavelength of excitation. We show in Ref. [22] the drastic difference between the dispersion of the enhanced fields in the core and the scattered fields just outside the shell.

These features represent an ideal condition to boost the naturally weak nonlinear effects of optical materials [18,19]. For this purpose, we propose to load a nonlinear Kerr material in the core with relative permittivity $\varepsilon_c = \varepsilon_L + \chi^{(3)}|E|^2$, $\varepsilon_L = 2.2$, and $\chi^{(3)} = 4.4 \times 10^{-20}\ \mathrm{m^2/V^2}$ [23], where $|E|$ is the magnitude of the mean value of the local complex electric field in the core of the nanoparticle, calculated by using full-wave Mie theory. Since the field distribution is largely dominated by the dipolar fields, the electric field in the core is uniformly enhanced and linearly polarized over the entire frequency range of interest, well justifying the adopted mean-value approximation. All-optical switching effects are expected in this configuration, as demonstrated in Refs. [24,25] for a plasmonic grating. Moreover, given the large field enhancement, relatively moderate input pumping intensities may be required.

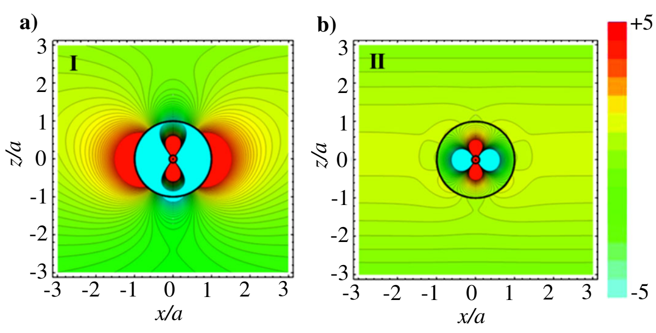


FIG. 2 (color online). Distribution of $E_x$ (snapshot in time) for (a) resonant scattering [point I in Fig. 1(c)] and (b) cloaking (point II) wavelengths. The impinging plane wave has unitary amplitude, and the electric field is linearly polarized along $x$, traveling from bottom to top. The aspect ratio is $\eta_c = 0.1$, similar to Fig. 1(c).

Figure 3 shows the normalized SCS for a relatively low input intensity $I_{\mathrm{in}} = 582\ \mathrm{MW/cm^2}$, compared with an uncloaked object with same nonlinear permittivity. Bistable scattering response is indeed found, with an abrupt switching effect at the design frequency spanning over 40 dB in total scattering reduction. In the plot, we also show the calculated unstable branch as a dotted line, which is a mathematical solution of the nonlinear problem, but it is not reachable from the two stable branches. The proposed device can swiftly change its scattering response from an efficient optical cloak to a resonant plasmonic scatterer as a function of the input intensity or of the wavelength of operation. Obviously, this sharp response spanning a fraction of a wavelength is achieved only in the lossless limit, and it may be hardly detected in a practical experiment. In case of realistic losses in the shell, however, the Fano-type resonance becomes broader and all-optical switching and bistable scattering response may still be obtained, with a lower scattering range, as discussed in Ref. [22]. Optimized cloaks based on this concept may make the range of bistability tunable to several degrees and applicable to a variety of nanodevices. The variation of the core nonlinear permittivity as a function of impinging intensity is shown in Ref. [22] for a specific wavelength of operation ($\lambda = 340.7$ nm), chosen at the center of the optical bistability region of Fig. 3. Bistable permittivity dispersion is observed, as expected, with a hysteresis curve characterized by a small nonlinear permittivity variation, corresponding to a relatively low input pumping intensity. This is associated with the fact that the nonlinear cloak sustains uniform, strongly enhanced fields inside the core

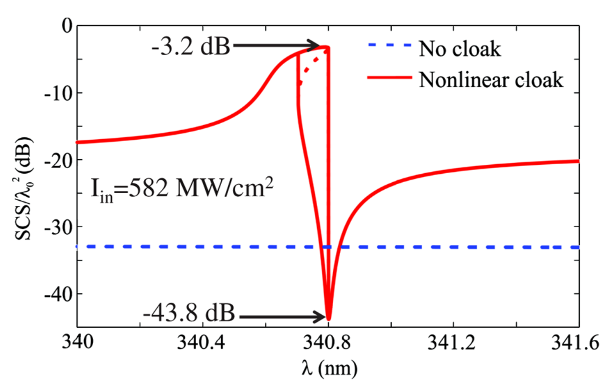


FIG. 3 (color online). Scattering response versus wavelength for nonlinear plasmonic cloaking operation [red (gray) line]. The device is pumped with moderate input intensity $I_{\mathrm{in}} = 582\ \mathrm{MW/cm^2}$. The unstable branch of the bistable curve is shown with a dotted red (gray) line. The scattering response of the uncloaked dielectric object corresponds to the dashed blue (dark gray) line. The nonlinear cloak can achieve more than 40 dB abrupt switching between resonant scattering and cloaking.

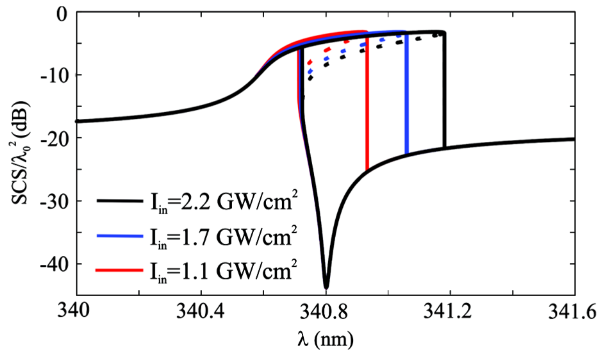


FIG. 4 (color online). Normalized scattering performance of a self-tunable bistable plasmonic cloak illuminated by three different input intensities: $I_{\text{in}} = 1.1\ \text{GW/cm}^2$ [red (gray) line], $I_{\text{in}} = 1.7\ \text{GW/cm}^2$ [blue (dark gray) line], and $I_{\text{in}} = 2.2\ \text{GW/cm}^2$ (black line). Longer switching wavelengths are obtained with higher input intensities.

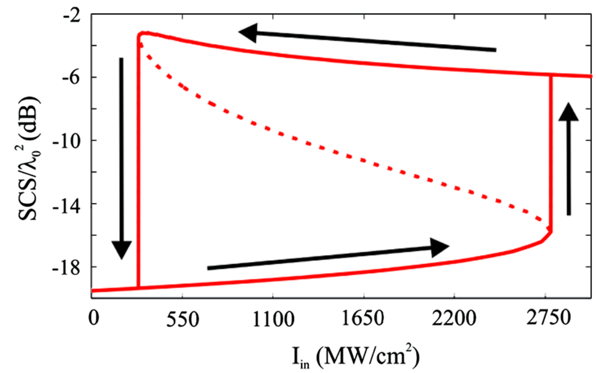


FIG. 5 (color online). Bistable normalized SCS as a function of input intensity for the nonlinear plasmonic cloak at $\lambda = 340.7$ nm. The unstable branch is shown with a dotted line. Strong hysteresis is obtained, ideal for all-optical switching applications.

(Fig. 2 and Ref. [22]), independent of the abrupt change of response in its scattering, directly leading to enhanced optical nonlinear effects and strong bistable response [19].

Figure 4 shows the hysteresis response of the nonlinear cloak for different input intensities: $I_{\text{in}} = 1.1\ \text{GW/cm}^2$ [red (gray) line], $I_{\text{in}} = 1.7\ \text{GW/cm}^2$ [blue (dark gray) line], and $I_{\text{in}} = 2.2\ \text{GW/cm}^2$ (black line). The bistability and overall scattering response of the nonlinear cloak gets broader for increased input intensity. Hence, the proposed structure may realize a nonlinear nanoswitch device, whose switching frequency is tunable with realistic input pumping intensity values. This tunability, combined with the all-optical switching properties discussed before, can directly lead to the design of nonlinear optical nanocircuit components [26] and new-generation tunable sensors.

Finally, Fig. 5 shows the hysteresis response of the proposed nonlinear plasmonic cloak as a function of impinging input intensity at the central wavelength $\lambda = 340.7$ nm. The broad hysteresis loop provides excellent functionality as a nanoswitch with high sensitivity to the flux intensity. When the intensity is increased, the normalized SCS remains almost constant with low values around $-18$ dB (lower branch of hysteresis curve). This can be considered the off-mode operation of the device, if we set an appropriate threshold $\text{SCS}_{\text{thr}} = -10$ dB. When the input intensities reach higher values than $I_{\text{in}} = 2750\ \text{MW/cm}^2$, the scattering performance switches to high normalized SCS values around $-6$ dB. However, when the optical intensity is decreased, the cloak operates in the on mode along the upper branch of the hysteresis curve. Only for much lower input intensity, below $I_{\text{in}} = 300\ \text{MW/cm}^2$, does the device go back to the off-mode operation. This example emphasizes the feasibility of the nonlinear cloak as an optical nanomemory.

In conclusion, we have proposed here the design and operation of a nonlinear plasmonic cloak. Its scattering response provides a unique Fano-like response for the total SCS, being detectable anywhere around the cloak and exhibiting inverse resonant dispersion compared to conventional Fano resonances. A sharp spectral response is achieved based only on interacting states of the dominant dipolar scattering mode, which makes this phenomenon purely symmetric, detectable in the total scattering of the particle. Moreover, it is not based on higher-order harmonics that require larger stored electromagnetic energy, which tend to be less scattering efficient in the presence of losses. Uniform, enhanced fields are obtained inside the device across the antiresonance, in both the bright and dark states, an ideal condition to boost naturally weak optical nonlinearities. When nonlinear elements are considered, a peculiar optical response is predicted: The nonlinear plasmonic cloak can exhibit an abrupt switch in visibility of up to 40 dB, rapidly varying between cloaking and resonant scattering. Strong bistability and tunability of the device can lead to interesting applications, such as low-intensity optical memories, switches, and sensitive tunable sensors. We may also consider planar arrays of these nanoparticles, whose collective response would present similarly sharp bistable features in the reflection and transmission coefficients, realizing integrated planarized optical nanodevices with strong, exotic nonlinear effects. As an example, the response of a dense periodic array of nonlinear cloaks with a square lattice with period $d = 2.25a$ is shown in Ref. [22].

The specific plasmonic design proposed in this Letter does not necessarily represent the optimal geometry to achieve tunable cloaking operation, but it should be viewed as an example to demonstrate the general concept of nonlinear "antiresonant cloaking" to "resonant scattering" mechanism, based on an alternative Fano mechanism based on dipole-dipole interaction to enhance nonlinear effects and sensitivity. Losses, as expected, have a detrimental effect on the resonant performance of the structure, especially for shorter wavelengths, as considered here, reducing the scattering peak and the hysteresis loop [22]. In addition, the broader bandwidths associated with lower $Q$ factors in the case of losses may require larger pumping intensities to trigger the switching effects. Still, the advantage of being based on purely dipolar fields may minimize the level of stored energy for a given linewidth, and optimized shapes and materials may allow shifting the operation to lower frequencies, for which metallic losses are less dominant and robust nonlinear response may be achieved. Still, even in the present design, after considering realistic silver losses as shown in Ref. [22], a similar scattering signature is obtained and the general features highlighted in this Letter can be observed by using realistic metals and pumping intensity levels.

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