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Sierpiński fractal plasmonic nanoantennas

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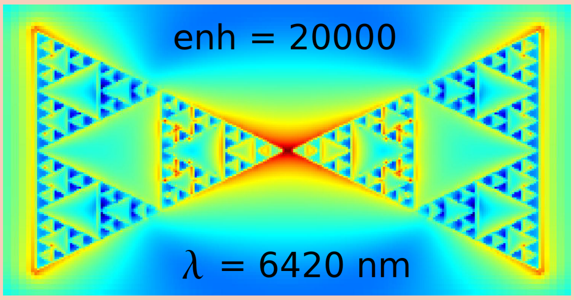
We propose plasmonic Sierpiński gasket, a self-replicating fractal, with structural elements spanning from $\sim$ 100 nm to $\sim$ 5 μm made by standard electron beam lithography (EBL), metal deposition, and lift-off sequence. Such structures demonstrate light field enhancement from visible to far-IR spectral range and can be scaled up towards THz band. Numerical simulations show that as the fractal order is increased, the optical extinction band broadens from the visible light towards far-IR, achieving a light field enhancement of more than four orders of magnitude in the nano-gap proximity. Such antennas are prospective for IR-THz filter, detection, and emission applications.



Fundamental mode simulation of Sierpiński fourth-order fractal gold nanoantenna with 10 nm nano-gap, made by EBL and lift-off.

1 Introduction Surface plasmon resonance in metallic subwavelength structures can be used at optical wavelengths to efficiently collect and retransmit light, and to guide it to specific surface locations through a strong electric field enhancement phenomenon known as as nanofocusing [1]. This process is affected by several factors, including electron crowding and back-reflections, causing strong current interest in its physical underpinnings. Moreover, the high field gradients on the metal surface pose a challenging simulation problem, as simulated enhancement can be affected by the used mesh and precision [2].

In this letter, we introduce, numerically simulate, and fabricate a novel kind of nanoantenna, with an optical response ranging from visible wavelengths and expanding towards the THz band, capable of achieving significantly high field enhancement in the nanogap on a very wide band, by exciting plasmon resonance modes in a self-replicating fractal metallic structure. A fractal-

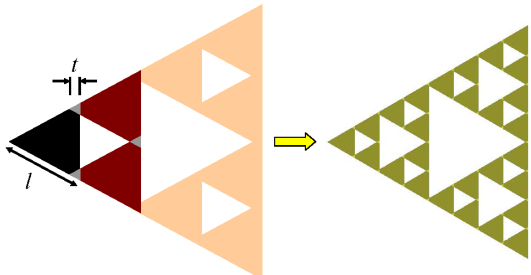


Figure 1 Evolution of elementary equilateral triangle of side length l (black) into self-similar Sierpiński gasket of first (dark red), second (pink), and third order (gold) with overlap t.

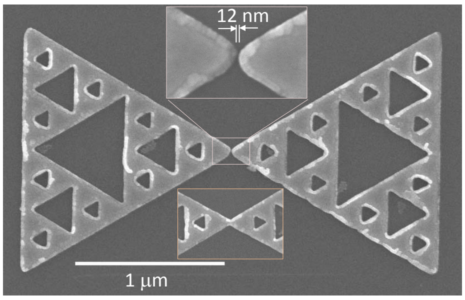


Figure 2 Scanning electron microscopy (SEM) image of the third-order Sierpiński nanoantenna, defined by EBL evaporation, and lift-off. Elementary triangle side length was 200 nm, and the top inset shows the nano-gap size, fixed at 12 nm. Triangle overlap is around 80 nm, and the gap can be controlled and even closed, as in the bottom inset where we have a 40-nm wide bridge.

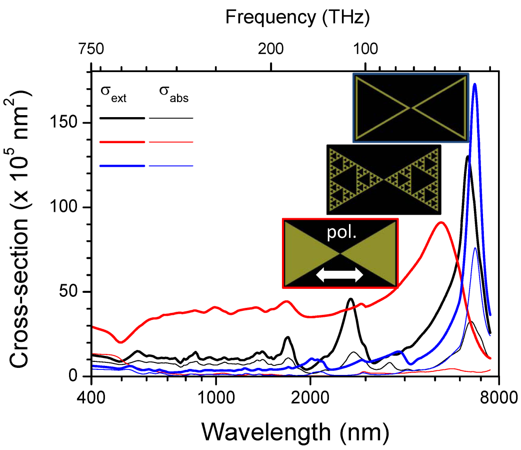


Figure 3 Numerically simulated extinction and absorption cross-sections of the fourth-order Sierpiński gold nanoantenna, compared with outline and solid bowtie, for linear polarization excitation as shown in the inset.

based 2D metamaterial has recently been shown to provide polarization-independent wide-angle subwavelength resolution imaging [3], while Sierpiński-type fractals are arising much interest for their broadband capabilities for applications such as optical trapping [4] and THz emission [5]. However, these structures, as designed, are either band-limited, which limits their usefulness, or of very large size up to 100 μm, which makes fabrication of tiny details difficult. Moreover, the photolithography methods used have limited resolution, reducing the feasible fractal order and thus the performance at the shorter wavelengths. We want to realize a compact structure, while pushing the Sierpiński order to high level by engineering nanometric details for the first time through EBL patterning in gold on a SiO_2 substrate, to control the size and features of the antenna down to nanometric precision. Such patterns are also required for electrochemical applications with a combined interdigitated contacts and surface enhanced Raman scattering (SERS) functionality [6].

The broadband nanoantenna is based on the Sierpiński gasket fractal, whose elementary building block is the 60° equilateral triangle. Since we are interested in preserving response up to visible wavelengths and incrementally broadening it to THz, we employ a slightly different construction method, starting with a basic triangle of height equal to 100 nm (side length $l = 115.5$ nm), which is able to provide coverage of the visible band in a bowtie configuration on a SiO_2 substrate. The triangle is then replicated twice at an offset to produce the first-order iteration shape, and the first-order shape is then replicated likewise to obtain the second-order shape, and so on, as shown in Fig. 1. The basic triangles are partially overlapped by a length $t = 5$ nm, in order to account for the finite size of the fabricated metal bridges, as shown in the title figure.

Here, we simulate optical properties of the nanoantennas prepared by EBL, where samples have been prepared in various orientations and fractal orders, of which an example is in Fig. 2, depicting a third-order fractal nanoantenna, with an obtained nano-gap width of 12 nm. The elementary triangle side length of this first-trial structure is of 200 nm, larger than the simulated one, but after process adjustment it will be possible to produce refined details and bring the size down to 100 nm or less. Overlap t also has a significantly higher value around 80 nm which needs correction in the final structure. This procedure permits to finely control the gap width, in fact the bottom inset in the same figure shows how it is possible to replace the gap with a bridge up to 40 nm wide, changing the boundary conditions and permitting to shift the plasmon resonances to even lower frequencies in the THz band. This kind of patterning is fast and accurate enough for this application, however if greater precision is required, further processing can be done via 3D nano-patterning by ion-beam lithography with nanometric precision [7]. The possibility of performing such fine and repeatable sample processing on large areas opens new horizons for nano-focusing in plasmonics.

2 Samples and simulations Samples are prepared by standard EBL, sputtering of of gold on 2-nm-thick Cr adhesion layer, and lift-off as described in [8]. 40-nm-tall gold nanostructures were deposited on silica glass. Nanoparticles patterns were characterized by scanning electron microscopy (SEM).

The optical response (cross-section and field enhancement factor) of the nano-antennas was simulated with a finite differences time domain (FDTD) numerical solver (Lumerical). Reticule gradation is employed to increase mesh accuracy in the areas of greatest field enhancement, especially in the central nano-gap. The nano-antenna is simulated in the band from 400 to 7500 nm (40 to 750 THz), divided in 1500-nm bands to achieve a 1 hour simulation for up to 0.5 ps propagation of the time-domain

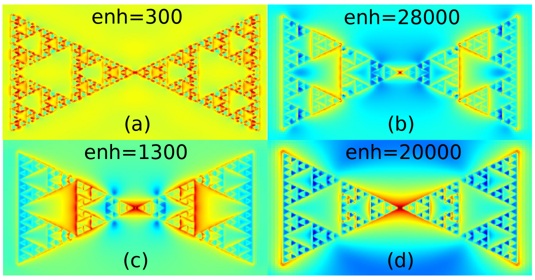


Figure 4 Electric field intensity distribution log-plot on the gold-silica interface for the fourth-order Sierpiński nanoantenna, at (a) 700 nm, (b) 2950 nm, (c) 3540 nm, and (d) 6420 nm.

broadband pulse. Materials are modeled by their complex permittivity values according to the Lumerical database.

3 Results and discussion To calculate the blocking surface offered by the gold nano-antenna, we calculate the dimension of the Sierpiński gasket by taking into account the introduced triangle overlap. In particular, being $A_0 = l^2\sqrt{3}/4$ the area of the elementary triangle and $A_t = t^2\sqrt{3}/3$ the area of the overlap triangle, the total area of the solid surface A_i at iteration i, is given by:

$$A_i = 2\left(A_0 3^i - A_t \sum_{k=1}^{i} 3^k\right) = 2A_0 3^i + 3A_t\left(1 - 3^i\right)$$

The fourth-order Sierpiński antenna as simulated has a length of 2.91 μm and an area $A_4 = 0.93$ μm^2. It is interesting to compare the performance of this structure with two other non-fractal structures, namely the solid gold bowtie antenna of the same footprint (area 2.42 μm^2), and a similar one that is made of only a 50-nm wide triangular solid circuit outlining the bowtie shape, which gives the minimum possible surface area of 0.48 μm^2.

Extinction and absorption cross-sections are compared in Fig. 3. The outline bowtie has a strong extinction peak around 6700 nm while settling down to values less than one-twentieth; in the solid structure circulation of electrons is less constrained, thus cross-section at lower wavelengths is 5 to 8 times higher, though the bandwidth is 20% less. Interestingly, both the non-fractal structures show very low absorbance at wavelengths below the main peak and, as the loss is mainly due to losses in material, this is linked to a predominance of wave scattering, while minimal field-enhancing plasmonic resonance is excited.

The fractal structure instead, shows several peaks linked to excited plasmon resonances, the main peak around 6300 nm being in-between the two bowties. Other peaks appear at 1700 nm and 2700 nm, the latter one being over the extinction values of the bowties, but interestingly the extinction remains consistent below 1500 nm, averaging around values that are 3 to 4 times the outline-bowtie and about one third of the solid one. The absorption cross-section is 4 to 5 times higher than that of bowties, indicating a complex distribution of plasmonic modes providing field enhancement up to visible range. An interesting aspect of this patterning process is the ability to finely control absorption and scattering bands of the nano-antennas.

By looking at the electric field intensity distribution on the gold-silica interface, several main plasmonic modes can be identified and their enhancement measured (Fig. 4). The fundamental one (Fig. 4(d)) at 6420 nm resonates on the whole fractal shape, with hotspots at the nanogap and at the outer corners for a maximum nanogap enhancement around 2×10^3. The inner structures support two modes, one less focused at 3540 nm (Fig. 4(c), enh. 1300), and one more focused at 2950 nm (Fig. 4(b), enh. 2.8×10^3). Below 1500 nm, energy distributes throughout the structure giving a large number of hotspots; enhancement is reduced but still significant, e.g., at 700 nm (Fig. 4(a), enh. 300).

The most remarkable feature of this structure is the ability to provide extremely strong field enhancement as it can "squeeze" a field with a wavelength in the order of 10 μm into a nano-gap whose size is in the order of 10 nm, suggesting that a proper structure scaling with increased fractal order can focus a 100 μm field (3 THz) in a 100 nm gap with comparable enhancement. Notice that the THz mode in Fig. 4(d) is indeed localized, but not very sensitive to the nano-gap size, as it is distributed on a quite larger area, so we can modify the structure by joining it with a bridge (Fig. 2) and push the resonance down to around 20 THz without compromising enhancement. This leads to further engineering of the fractal shape in order to increase the number of supported modes for even wider band coverage to reach down to the low end of the THz band.

4 Conclusions We numerically showed that EBL-patterning of gold on silica can be used to realize novel broadband Sierpiński fractal nanoantennas with strong nanogap field enhancement from 40 THz to visible. The fabrication process with 10-20 nm precision can be cost-effective for production of nanoantennas, with promising applications in SERS sensing. This numerical study leads the way for upcoming trials of Sierpiński fractal nanoantennas fabrication for ultra-wideband THz-to-visible applications, whose technical feasibility is demonstrated.

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