U-Shaped Switches for Optical Information Processing at the Nanoscale

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Fully light-based circuits are becoming a realistic possibility due to the recent advances in metamaterials. The latter are artificial materials engineered for displaying unusual optical properties, such as negative refractive index,[1] extraordinary transmission,[2] superlensing, cloaking etc.[3] In an optical nanocircuit, metamaterials could take the place of standard electronic components.[4–6] In such devices, light waves would be used instead of electrons. The possibility arises from the fact that light waves can couple to collective excitations of electrons at the surfaces of metallic nanostructures, a property referred to as surface plasmon resonance.

Because these optically induced resonances occur at the surfaces and interfaces of the nanostructures, they can readily be investigated with a surface- and interface-specific optical technique, such as second-harmonic generation (SHG). SHG is a nonlinear optical technique that, within the dipole approximation, is forbidden in materials with a center of symmetry. Consequently, SHG is highly sensitive to regions with broken symmetry, such as surfaces (or interfaces), and it has been successfully applied to the study of plasmonic nanomaterials with different geometries.[7–10]

Of particular interest here is the U-shaped nanostructure geometry. This type of structure has attracted a lot of attention due to its particular resonances,[11] magnetism,[12] and possible negative index of refraction.[13] The letter U has two extremities that are alike (same size, parallel) and yet different (due to the connection). The similarity and distinctiveness of these extremities could perhaps be put to use in interacting with an optical nanocircuit. Indeed, while optical nanocircuit equivalents for simple electronic components, such as the resistor or the capacitor, have already been proposed, information processing requires control over the flow of light in the optical nanocircuit—a switch.

Herein, we report that maxima of plasmonic currents occur at the two extremities of U-shaped nanostructures. These plasmonic currents can be turned ON and OFF, separately or simultaneously, depending on the polarization state of an incoming light beam (see **Figure 1**a). Upon coupling the extremities (or outputs) of the letter U to the entrances of an optical nanocircuit, the U-shaped nanostructures could act as a switch that is regulated by the polarization of light, for channeling plasmonic currents and, therefore, information (see Figure 1b). Additionally, the switch could act as a gate that enables or disables plasmonic current in an optical circuit by acting on circuit elements that are sensitive to heat, light, or strong local fields (such as photodiodes, thermal switches, etc.; see Figure 1c). A particular strength of our work is that the plasmonic field enhancements are mapped both experimentally and theoretically, with a remarkable agreement between the two methods. At variance with previous results showing asymmetric redistribution of plasmonic current,[14] our SHG data do not represent a snapshot and the asymmetry does not reverse with the frequency. Consequently, our particular structures constitute a practical switch.

The sample under investigation consists of a periodic array of five nanostructures having different geometries (see **Figure 2**a). All the nanostructures are 200 nm apart and consist of gold lines that are 200 nm wide and 25 nm thick. Of interest here are the U-shaped nanostructures, highlighted in yellow. These structures are 600 nm long and 400 nm wide. The samples are prepared on a SiO_2(100 nm)/Si(001) substrate by electron-beam lithography, with a double resist layer of poly(methyl methacrylate) (PMMA/co-MMA) to facilitate the subsequent lift-off. The Au deposition is carried out in a molecular beam epitaxial system at a pressure of $\approx 5 \times 10^{-9}$ mbar and a rate of ≈ 0.2 Å s^{-1}. Lift-off is then performed with boiling acetone. To check the quality of the deposited nanostructures, scanning electron microscopy (SEM) is performed with a commercial instrument (Jeol, JSM5600).

The presence of plasmonic hot spots at the outputs of the switch is directly evidenced by means of SHG microscopy

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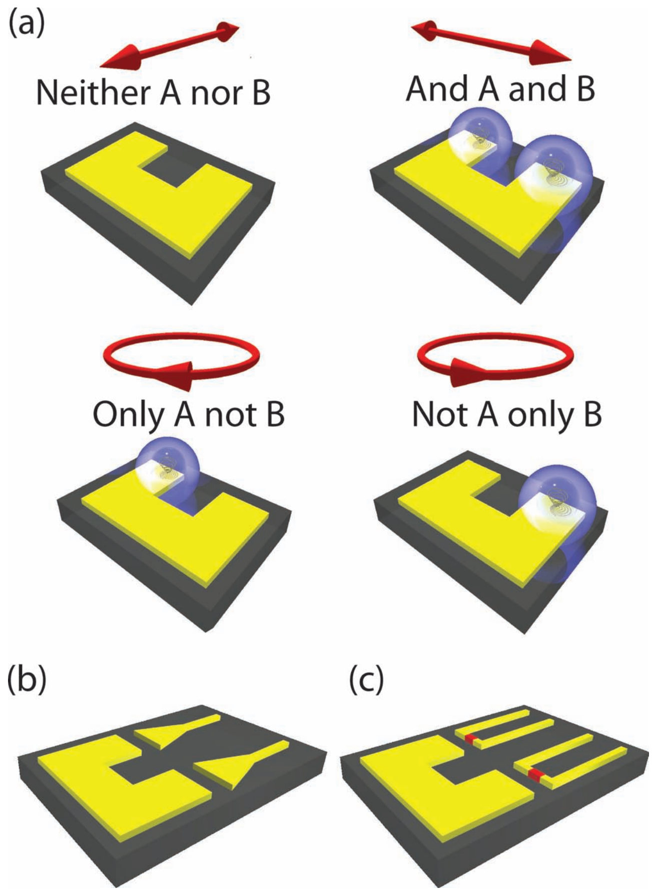

Figure 1. Plasmonic switch for optical information processing at the nanometer scale. a) Depending on the polarization state of the incoming light (λ = 800 nm), the two branches (outputs A and B) of a golden U-shaped nanostructure give rise to localized second harmonic sources (λ = 400 nm), or hot spots, that are due to local field enhancements. The nanostructure is 600 nm long, 400 nm wide, and 25 nm thick. A and B are both 200 nm wide. b,c) Schematic diagrams of the basic principles for interaction with a circuit. More specifically, in (b) the extremities of the U-shaped nanostructure could provide entrance points to an optical circuit. In (c), the switch could act as a gate that enables or disables the current in an optical circuit by acting on a photodiode, thermal switch, etc. (represented in red).

images. These images are collected with a confocal laser scanning microscope[15] (Zeiss, LSM 510) using a femtosecond pulsed Ti:sapphire laser (Newport, DeepSee Mai-Tai). The 800 nm ultrafast laser pulses are dispersion-compensated for the broadening that occurs in the objective (Zeiss, 100×/1.46 oil immersion).

Both the quality of the sample and the settings on the SHG microscope are verified by depositing a reference nanopattern and comparing its SHG response to previous studies. For this reason, the right half-side of the sample area in Figure 2a is imprinted with square-type spirals, which were investigated in the past.[16,17] Figure 2b–e shows the SHG images corresponding to different states of the incoming polarization. All images show hot spots, whereby the color-coded intensities increase from purple through blue, green, yellow, and red to white. The position of these hot spots is revealed through superimposing the SHG images upon the nanostructure geometry. For both linearly polarized (Figure 2b,c) and circularly polarized incoming light (Figure 2d,e), the reference hot-spot patterns of the square-type spirals is in good agreement with previous observations.

The polarization at the second-harmonic frequency can be written as:[18]

$$P_i(2\omega) \propto P_i^{D}(2\omega) + P_i^{Q}(2\omega) = \chi_{ijk}^{(2)} : E_j(\omega)E_k(\omega) + \chi_{ijkl}^{(3)} \vdots E_j(\omega)\nabla_k E_l(\omega) \quad (1)$$

where $\chi_{ijk}^{(2)}$ and $\chi_{ijkl}^{(3)}$ are second- and third-rank susceptibility tensors, respectively, while i, j, and k represent any of the Cartesian coordinates X, Y, and Z. The coordinate system is oriented so that X and Y are in the plane of the sample, while Z is perpendicular to the sample. The first term in Equation (1) is the surface-specific electric dipole contribution, indicated by the index D. The second term in Equation (1) includes the bulk-specific electric quadrupole and magnetic dipole contributions, indicated by the index Q.

It can be shown that the intensity at the second harmonic can be expressed as:[19]

$$I(2\omega) \propto \left| L(2\omega)\, L^2(\omega)\, \mathbf{P}(2\omega) \right|^2 \quad (2)$$

where $L(\omega)$ and $L(2\omega)$ represent the local field factors for the fundamental frequency and contain the contributions from plasmonic excitations, such as those observed in our nanostructures.

In gold, the surface dipole contributions are typically much larger than those of the bulk quadrupoles. Additionally, the local field factors can be greatly enhanced due to surface plasmon resonances. For these reasons, we believe that the surface contributions are dominant in the SHG signal that we observe.

For the purpose of clarity, in Figure 2 and **Figure 3** the extremities of the U-shaped nanostructures are labeled A and B, while the presence or absence of SHG hot spots is emphasized by juxtaposing full or empty white circles, respectively. These white circles indicate the ON/OFF states of the switch as a function of the light's polarization. The latter is studied in four distinct cases. First, Figure 2b indicates that, upon applying horizontal linearly polarized light, no bright hot spots can be seen (neither A nor B). Nevertheless, careful examination reveals a purple spot that seems to be positioned at the center of the whole nanostructure. Next, in Figure 2c, rotating the direction of linearly polarized light by 90° results in two hot spots, each appearing at one of the extremities of the U-shaped nanostructure (and A and B). The slight variation of intensity of the hot spots can be attributed to small fabrication defects, to which the SHG signal is known to be very sensitive.[20] Subsequently, in Figure 2d, for left-hand circularly polarized light, there is a single hot spot situated at the B extremity (not A, only B). And finally, in Figure 2e, for right-hand circularly polarized light, there is a single hot spot situated at the A extremity (only A, not B). These results demonstrate that the SHG hot spots exhibit four distinct

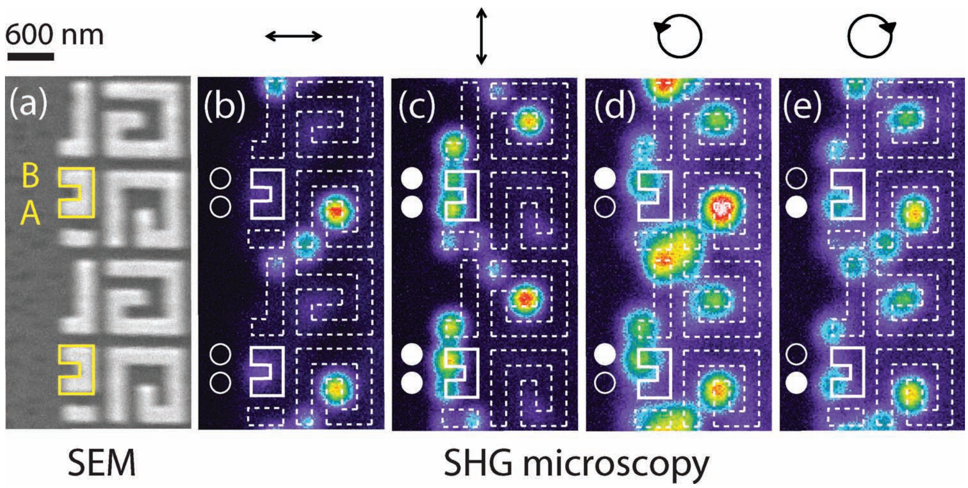

Figure 2. SHG microscopy reveals four types of local field enhancements in U-shaped gold nanostructures, depending on the polarization state of the incoming light. a) SEM image of the nanostructures. The U-shaped nanoelement is highlighted in yellow for clarity. SHG microscopy shows the hot spots due to local field enhancement by plasmons, for the linearly polarized light—horizontal in (b) and vertical in (c)—and for the circularly polarized light—left-hand in (d) and right-hand in (e)—as seen from the source. The color-coded intensities increase from purple through blue, green, yellow, and red to white. The locations of the SHG hot spots are displayed by superposing them on the SEM images. For clarity, the presence or absence of hot spots on the U-shaped nanostructures is indicated by full or empty white circles, respectively.

logical states depending on the polarization of the incoming light, but are these SHG hot spots representative of the plasmons at the fundamental frequency or are they solely related to nonlinear optical behavior?

In Figure 3, rigorous numerical simulations of the total local electric currents in the U-shaped nanostructures are displayed. The simulations are carried out with MAGMAS, an electromagnetic solver originally developed at the Katholieke Universiteit Leuven for problems in the microwave and millimeter wave bands. The solver has been extended to include the special features of plasmonic nanotechnology: (near) optical frequencies and strongly dispersive materials. Using the integral equation formulation,[21] for topologies embedded within a multilayered structure, it solves the linear Maxwell equations. We favor the use of MAGMAS because our knowledge and understanding of the code allow us to customize all conditions for the simulations. In Figure 3, the calculations were performed taking into account the substrate and the surrounding dielectric—air—for incoming light waves at 800 nm. Although the SHG experiments were conducted with very high numerical aperture, it was shown that this numerical aperture has no noticeable effect on the results;[22] consequently, the light wave used for the simulations was plane-polarized. For all the previously mentioned polarization states, the electric currents at the surface of the nanostructures are shown. The color-coded intensities increase from blue through green and yellow to red. First, Figure 3a indicates that, upon applying horizontal linearly polarized light, no maximum of total currents is present at the extremities of the U-shaped nanostructure (neither A nor B). Nevertheless, two small maxima can be observed at the inside corners of the nanostructure. These maxima can explain the purple spot in the center of the U-shaped nanostructure in Figure 2b. Next, for vertical linearly polarized light, there are two large maxima of local currents at the extremities of the U-shaped nanostructures (and A and B; see Figure 3b). Subsequently, as shown in Figure 3c, for left-hand circularly polarized

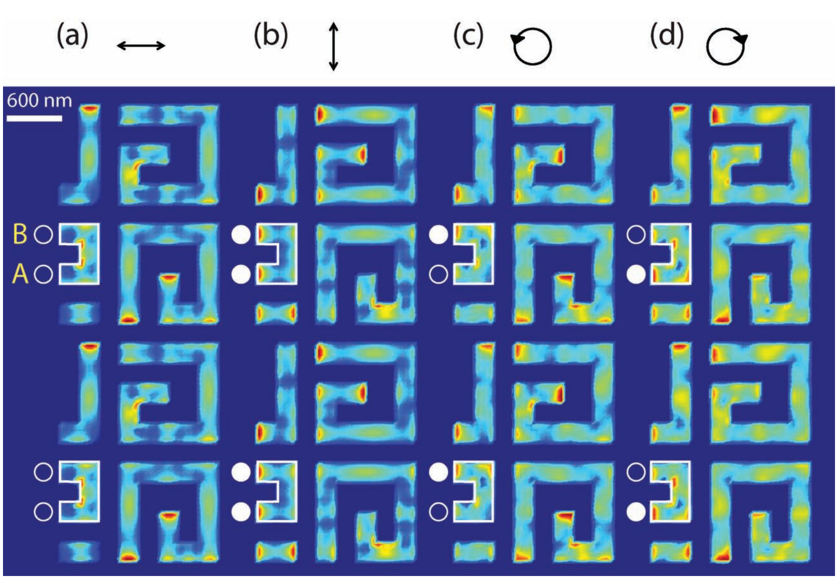

Figure 3. Theoretical simulations of the total electrical currents in the U-shaped nanostructures. a) For vertical linearly polarized light there is no local current at the outputs of the switch. b) For horizontal linearly polarized light, the local current is maximum at both the A and B outputs. c) For left-hand circularly polarized light, the local current is maximum at output B. d) For right-hand circularly polarized light, the local current is maximum at output A. The color-coded intensities increase from blue through green and yellow to red.

light, there is a single large maximum of the local current, situated at the B extremity (not A, only B). And finally, in Figure 3d, we see that for right-hand circularly polarized light, there is a single large maximum of the local current, situated at the A extremity (only A, not B). These numerical simulations of the linear optical behavior are in good agreement with the SHG microscopy results.

In conclusion, we have applied SHG microscopy to investigate the plasmonic behavior of U-shaped nanostructures made of gold. The SHG images reveal the occurrence of hot spots at the extremities of the letter U, which can each be switched ON and OFF depending on the polarization state of the incident light. Based on the very good agreement between experimental data and numerical simulations, the hot spots are attributed to maxima of plasmonic currents. We believe that these currents could be coupled to optical nanocircuits, whereby the U-shaped nanostructures would constitute a switch for channeling information depending on the polarization of light.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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