

Supporting Information

Color Routing via Cross-Polarized Detuned Plasmonic Nanoantennas in Large Area Metasurfaces

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TILTED OPTICAL NANOANTENNAS FABRICATION

A soda-lime glass substrate (20 × 20 × 2 mm) is repeatedly rinsed in ethanol and acetone. The sample is then placed in a custom-made vacuum chamber and irradiated with an 800 eV low energy defocused Ar⁺ ion beam (gas purity N5.0). A biased tungsten filament avoids charge build-up through thermionic electron emission. The ion beam illuminates the glass surface at an incident angle of $\theta = 30^\circ$ with respect to its normal. The ion fluence corresponds to 1.4×10^{19} ions/cm² and the glass temperature is fixed at about 680 K during the Ion Beam Sputtering (IBS) process. After the rippled pattern is formed on the glass surface, thermal Au deposition is performed on the rippled facets at a glancing angle $\theta = 55^\circ$ with respect to the flat sample normal. The Au beam directly illuminates the glass facets tilted at $+35^\circ$ while the opposite facets are completely shadowed. By means of a calibrated quartz microbalance, the thickness h of the Au stripes can be

evaluated by basic geometrical arguments given the Au thickness (h_0) deposited on a flat surface facing the crucible at normal incidence and the average slope of the illuminated facet measured with AFM as $h = h_0 \times \cos(55^\circ - 35^\circ)$. The sample is then put in a custom-made RF sputtering chamber where a layer of SiO₂ is conformally grown all over the surface using a 2" fused silica target. The silica layer thickness was monitored by means of a calibrated quartz microbalance. The RF sputtering experiment is run in Argon atmosphere at a power $P = 60$ W, sample-target distance $d = 8.5$ cm and total pressure of about $P = 7 \times 10^{-2}$ mbar. Finally, Ag stripes are confined on the rippled facets tilted at -50° , now coated with a conformal SiO_x layer, by using the same strategy and arguments already described for the Au ones.

MORPHOLOGICAL CHARACTERIZATION

The rippled glass template morphology was characterized by means of an atomic force microscope (Nanosurf S Mobile), running in tapping mode. The average periodicity and slope of the glass ripples were computed from the statistical analysis of AFM topographies by means of WSxM software (Fig. S11). Top view and cross section back scattered electrons images of the bimetallic nanoantennas array were acquired by means of a scanning electron microscope (Hitachi VP-SEM SU3500), operating in the 10-15 kV accelerating voltage range. Statistical analysis was performed on the SEM images by means of ImageJ software in order to evaluate the average antennas width and average antennas and silica gap thicknesses.

Fig. S1a shows a typical AFM topography of the self-organized nanostructured glass template. The template average periodicity λ is estimated from the real space distance between the maximum and the secondary neighboring peaks in the 2D self-correlation function (Figs. S1b and S1c).

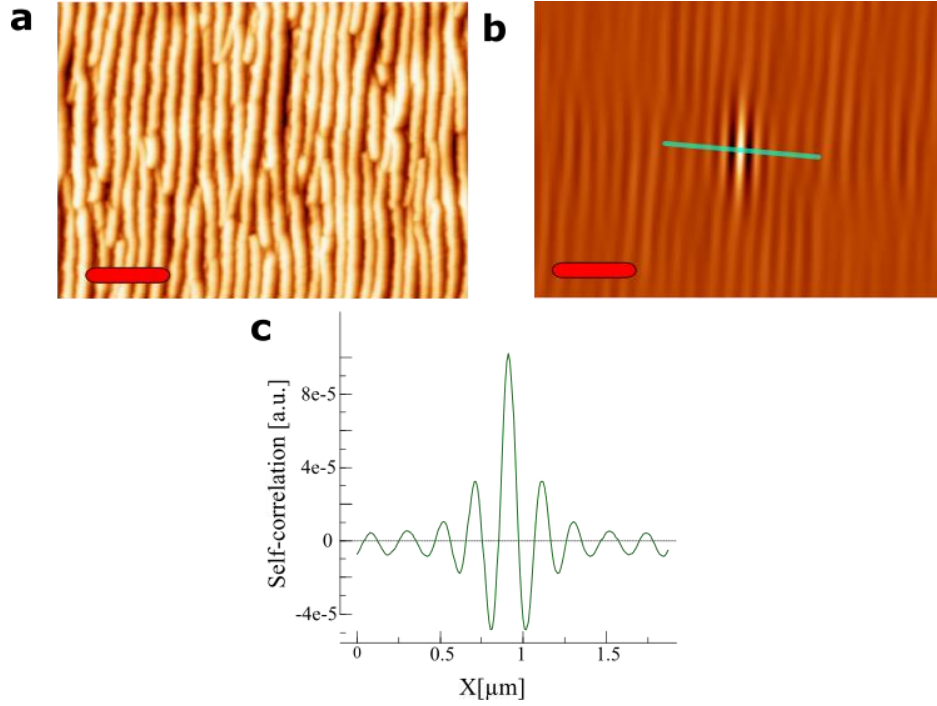


Figure S1: (a) AFM topography, (b) self-correlation function, (c) line profile (bottom panel) of the self-correlation function, extracted in correspondence of the green line in (b). The red scale bars correspond to $1\ \mu\text{m}$.

It's worth to note how the 2D self-correlation function of the rippled patterns rapidly decays to negligible values away from the central maxima, as the pattern loses its morphological coherence within 2-3 unit cells. This prevents the rippled glass template, and consequently the nanoantennas array confined on it, from showing grating optical effects which would lead to a more complex engineering of the color routing properties of our self-organized large area platform.

NUMERICAL OPTICAL MODEL

For the numerical analysis of the nanostrip antennas we employed a commercial software (Comsol Multiphysics 5.3), implementing the full-vectorial finite element method for scattered field formalism in two dimensions. We assumed a circular computational domain with 500 nm radius, surrounded by perfectly matched layers (PML) with scattering boundary conditions. The effective

environment approximation was assumed (in accord with, e.g., Refs. 1,2) by embedding the nanostrips into a homogeneous dielectric medium with non-dispersive and lossless permittivity.

The sketch of the bimetallic nanoantenna is shown in Fig. S2a. To avoid numerical artifacts, the vertices of the Au and Ag nanostrips have been rounded with 5 nm and 15 nm radius of curvature, respectively. The FEM mesh was accordingly defined so to resolve these radii with at least 5 elements. For the dielectric domain we set a maximum mesh element size corresponding to $\lambda/(5\sqrt{\epsilon_{eff}})$, with λ the optical wavelength in vacuum and $\epsilon_{eff} = 2.05$ the effective permittivity of the dielectric. The latter was estimated according to the so-called *effective environment approximation* as the mean value between the permittivity of the silica substrate (2.13 and air), corrected with a further increase (of ~ 0.48 , fitted by matching the spectral position of the plasmonic resonances) to take into account the conformal non stoichiometric silica spacer in-between the two nanostrips.

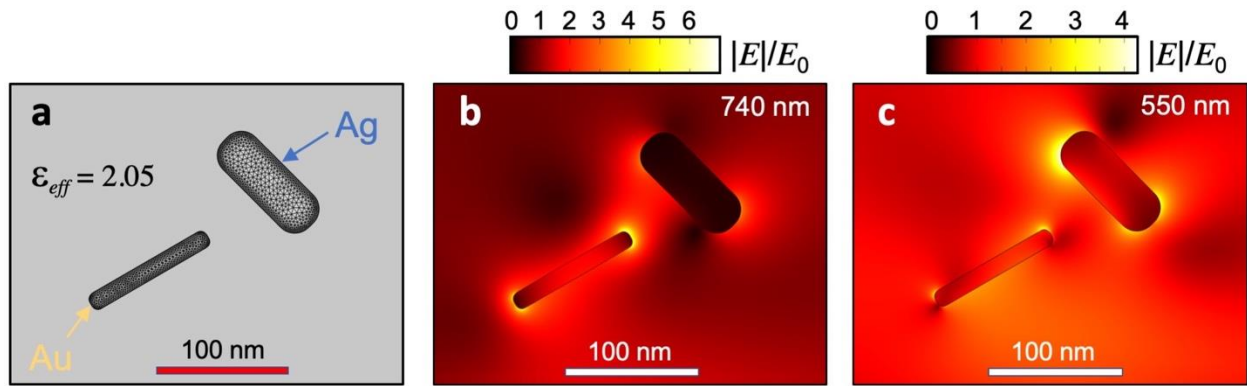


Figure S2: (a) Sketch of the simulated 2D geometry (also showing FEM mesh elements, only in the nanowires for better reading). (b)-(c) Simulated near field enhancement at (b) 750 nm and (c) 550 nm, which approximately correspond, respectively, to the dip and peak wavelengths in the simulated directivity spectra in the bottom panel of Fig. 4c (see main text).

Starting from the scattered electric $\mathbf{E}_S$ and magnetic $\mathbf{H}_S$ vector fields (numerically solved for as a function of λ), the total extinction cross-section is computed as $\sigma_E = \sigma_A + \sigma_S$, with σ_A and σ_S the total absorption and scattering cross-section spectra, respectively given by:

$$\sigma_A(\lambda) = \frac{\pi c \epsilon_0 \epsilon''_m(\lambda)}{\lambda I_0} \iint_{metal} |\mathbf{E}(\mathbf{r}, \lambda)|^2 dS$$

$$\sigma_S(\lambda) = \frac{1}{2I_0} \int_{\Sigma} Real\{\mathbf{E}_S(\mathbf{r}, \lambda) \times \mathbf{H}_S^*(\mathbf{r}, \lambda)\} \cdot \mathbf{n} dl$$

In above formulas, c is the speed of light in vacuum, ϵ_0 is the vacuum permittivity, Σ is a circle surrounding the nanoscatterers, and $\mathbf{n}$ is the outward pointing normal vector of the circle. Note that since our numerical model is in 2D, the cross-sections above detailed are lengths and not areas, and the intensity I_0 of the incident plane wave, is thus measured in W/m.

For the metal permittivity spectrum $\epsilon_m(\lambda) = \epsilon'_m(\lambda) + i \epsilon''_m(\lambda)$, we assumed the analytical model provided in Ref. 3 for Au, and in Ref. 4 for Ag, fitted on Johnson and Christy database. Note that, in order to mimick the inhomogenous broadening due to the dispersion of size (or periodicity) in our samples, we assumed an effective Drude damping parameter Γ as large as 5 times the ideal value Γ_0 reported in Refs. 3,4. This is in agreement with other studies on poly-dispersed plasmonic systems (see e.g. Refs. 5,6).

With the total extinction cross-section at hand, the transmittance of the sample at normal incidence is estimated as following:

$$T(\lambda) = \exp\left[-\frac{\sigma_E(\lambda)}{\eta L}\right]$$

where $L = 200$ nm is the measured average periodicity of the sample and η a dimensionless fitting parameter of the order of 1 (in our simulations we set $\eta = 0.45$).

Concerning the far-field scattering patterns, we employed the FAR-FIELD procedure in Comsol, implementing the Stratton-Chu formulas (see, e.g., Ref. 7) using Σ as the aperture enclosing our 2D antennas (either being the Ag or Au monomer, or the Ag-Au dimer).

OPTICAL CHARACTERIZATION

VIS-NIR extinction measurements were performed at normal incidence using a halogen-deuterium compensated lamp (DH-2000-BAL, Mikropak) as source and a solid-state spectrometer (HR4000, Ocean Optics), operating in the wavelength range 300–1100 nm, as detector. Scattering spectra were acquired by employing a custom made scatterometer which can collect light as a continuous function of the azimuthal angle, for a fixed selection of polar angles. The sample was illuminated from the bare flat glass side with a Visible and Near-Infra-Red broadband laser source (SuperK COMPACT by NKT Photonics) pulsed at high frequency (~ 10 kHz) in the super-continuum regime, to have a stronger signal intensity. A fiber coupled spectrometer Ocean Optics HR4000 was again used as detector. For both extinction and scattering measurements, the light source was linearly polarized to investigate the anisotropic optical properties of the nanoantennas array.

In Fig. S3 we show digital camera images of a fragment of a complete Au-Ag dimer covered sample, looked upon from the Ag side (Fig. S3a) and Au side (Fig.S3b) when illuminated from below by a white lamp. The two digital pictures were acquired with the same aperture and exposure parameters. Fig. S3 clearly shows the passive, macroscopic color routing action taking place over the whole sample.

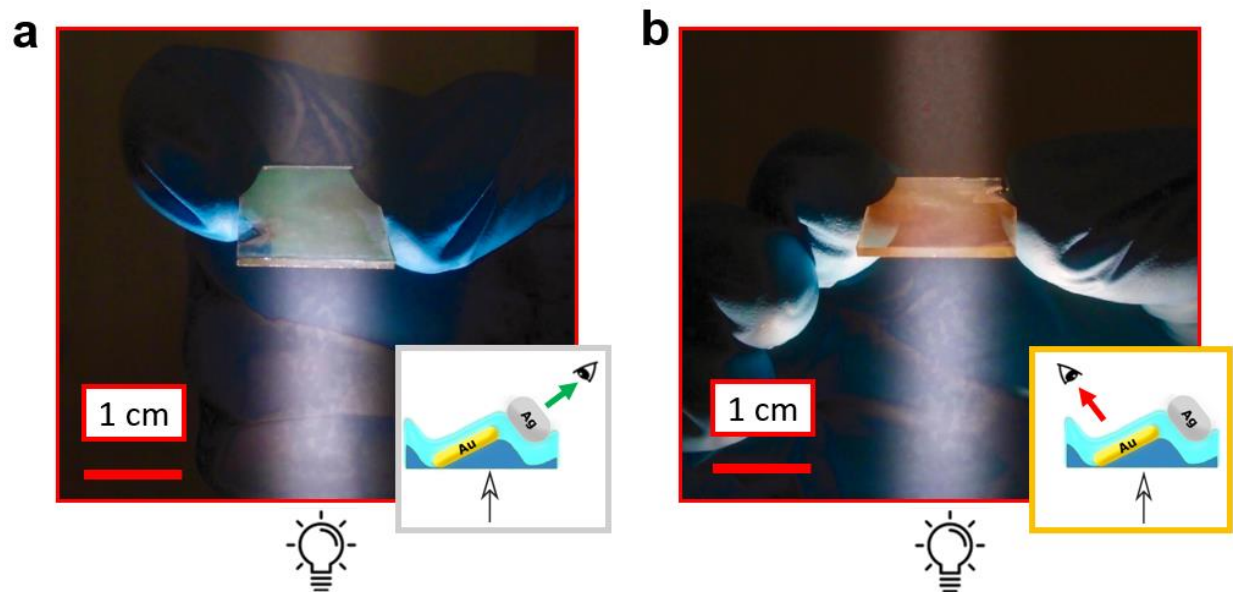


Figure S3: Pictures of a Au-Ag dimer sample fragment illuminated from below by a white lamp, looked upon from the Ag side (a) and Au side using the same acquisition conditions (aperture $F\ 3.5$, exposure time $1/250\ s$), (b). The insets present a sketch of the picture acquisition geometry. The incoming white light has been artistically rendered for clarity.

In Fig. S4 we show the measured directivity data for the Au, Ag and Au/Ag NSA reported in the main manuscript in Fig. 4a-b-c respectively, but using the same dB scale for all the panels. Fig. S4 evidences how the signal-to-noise ratio is similar for all the three different considered configurations when the directivity is not steeply changing with wavelength.

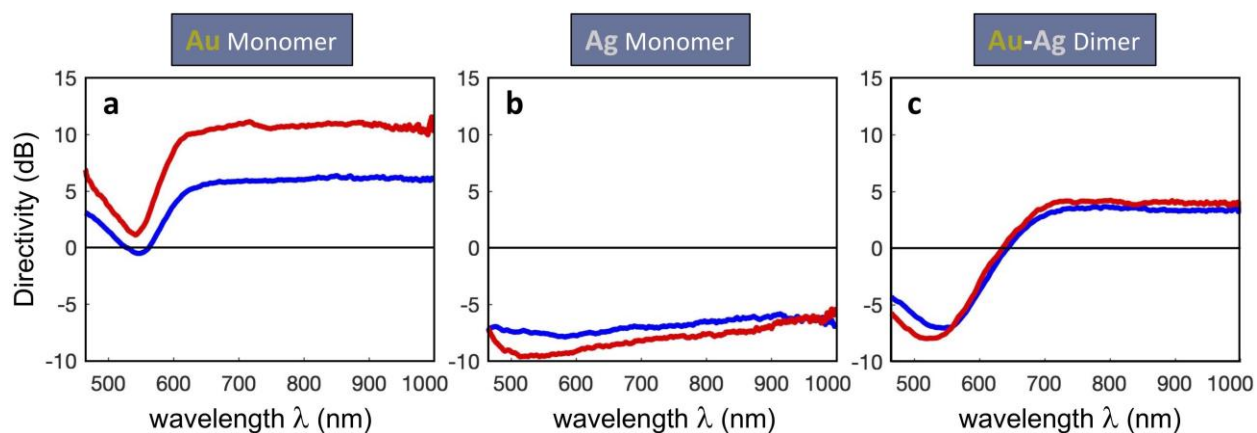


Figure S4: Measured (a, b, c) directivities at two different polar angles ($\Theta = 30^\circ$ in blue, $\Theta = 50^\circ$ in red) for: (a) Au NSA monomer, (b) Ag NSA monomer and (c) Au-Ag NSA dimer configurations, respectively.

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