

Supporting information

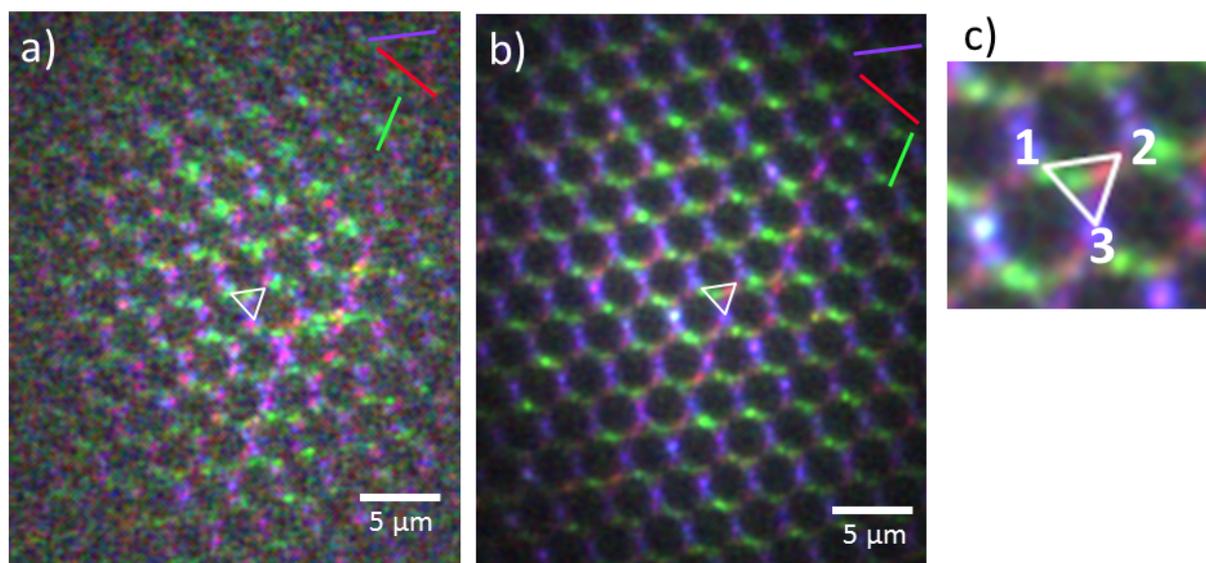


Figure S11: Merged images of three different a) SHG-images and b) 2PF images, respectively, each taken with a different plane of polarization of the incident light. The plane of polarization of the blue, red and green images are depicted by a line on the image in these respective colours, and c) an inset of b). As a guide to the eye, the white triangle corresponds in each of the three images. The images are taken such that each pixel of the CCD-camera detector depicts the averaged measured intensity of that pixel and its four neighbouring pixels. This leads to a lower spatial resolution, and a higher signal-to-noise ratio.

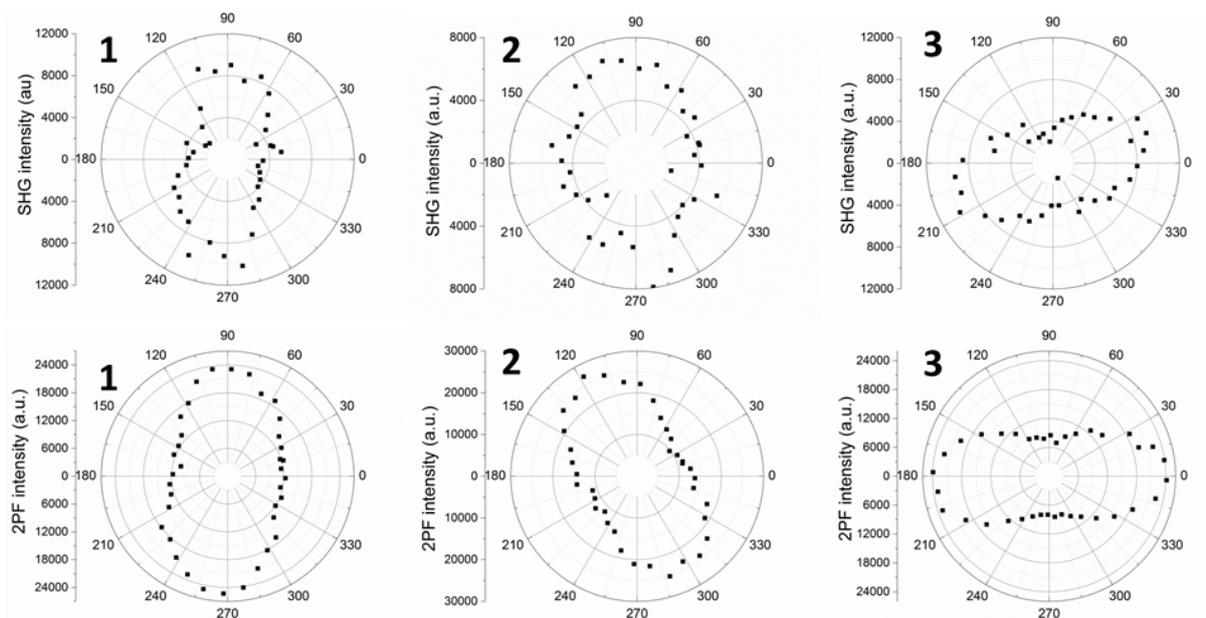


Fig. S12: SHG intensity (top) and 2PF intensity (bottom) in function of the plane of polarization of the incident light. The numbers assign to of which tip of the nanotriangle in figure S11 the SHG or 2PF intensity is shown.

Theoretical simulations

We provide information on the structure of the gold triangles for the simulations. Fig. S13 shows the gold triangles laying on the plane of the substrate (grey plane). The basis of the triangles is shaped via the used mask spheres of 3000 nm diameter, as drawn in blue on the grey plane. The corners are then smoothed to avoid field singularities at the tips. The vertical 40 nm gold thickness is grown following a spherical shape. Note that we design the smallest possible unit cell using Perfect Electric Conductor for boundary planes perpendicular to the x-axis and Perfect Magnetic Conductor for the boundary planes perpendicular to the y-axis for the electric field polarized along x. The inverse condition is employed for the y polarization.

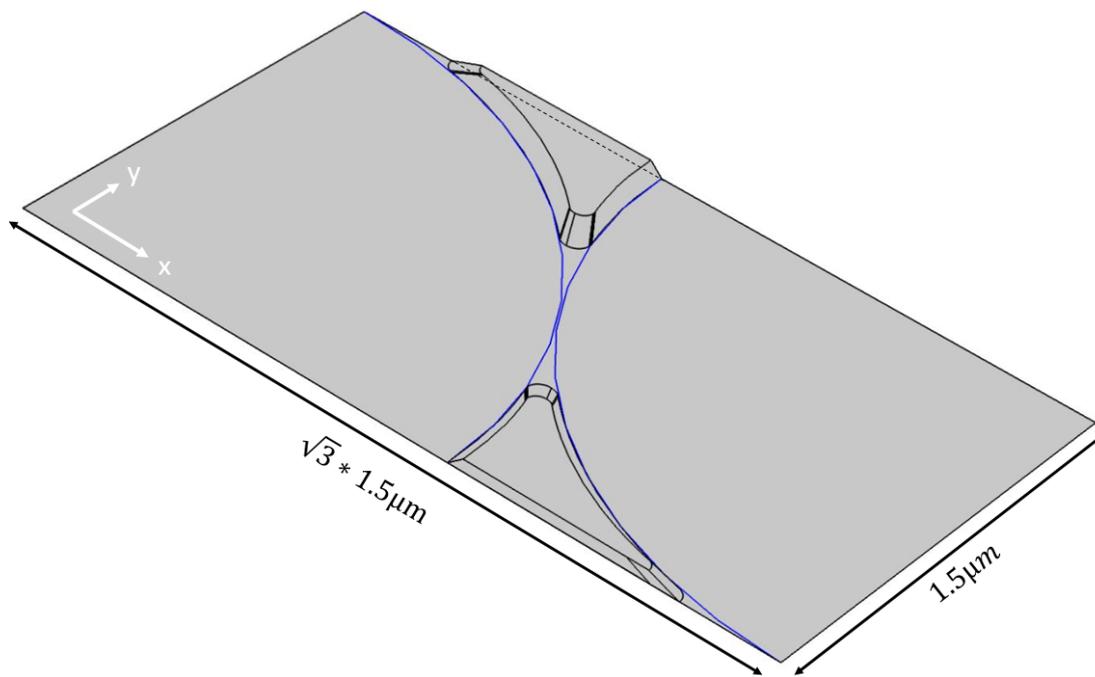


Fig. S13. Structure of the gold triangles in the theoretical simulations lying on the plane of the substrate.

Fig. S14 provides the fundamental dipole-like field profile. We plot the norm of the enhanced electric field (total field over incident field) 20 nm above the substrate at wavelength $2.55\mu\text{m}$. The two polarizations are represented and one clearly sees that the largest fields are generated on the triangular tips that are aligned parallel to the polarization direction. The magnitude of the field is about four times the resonance of the higher order mode depicted in Fig.3. Note that the power of the second harmonic generated wave should be proportional to the square of the field.

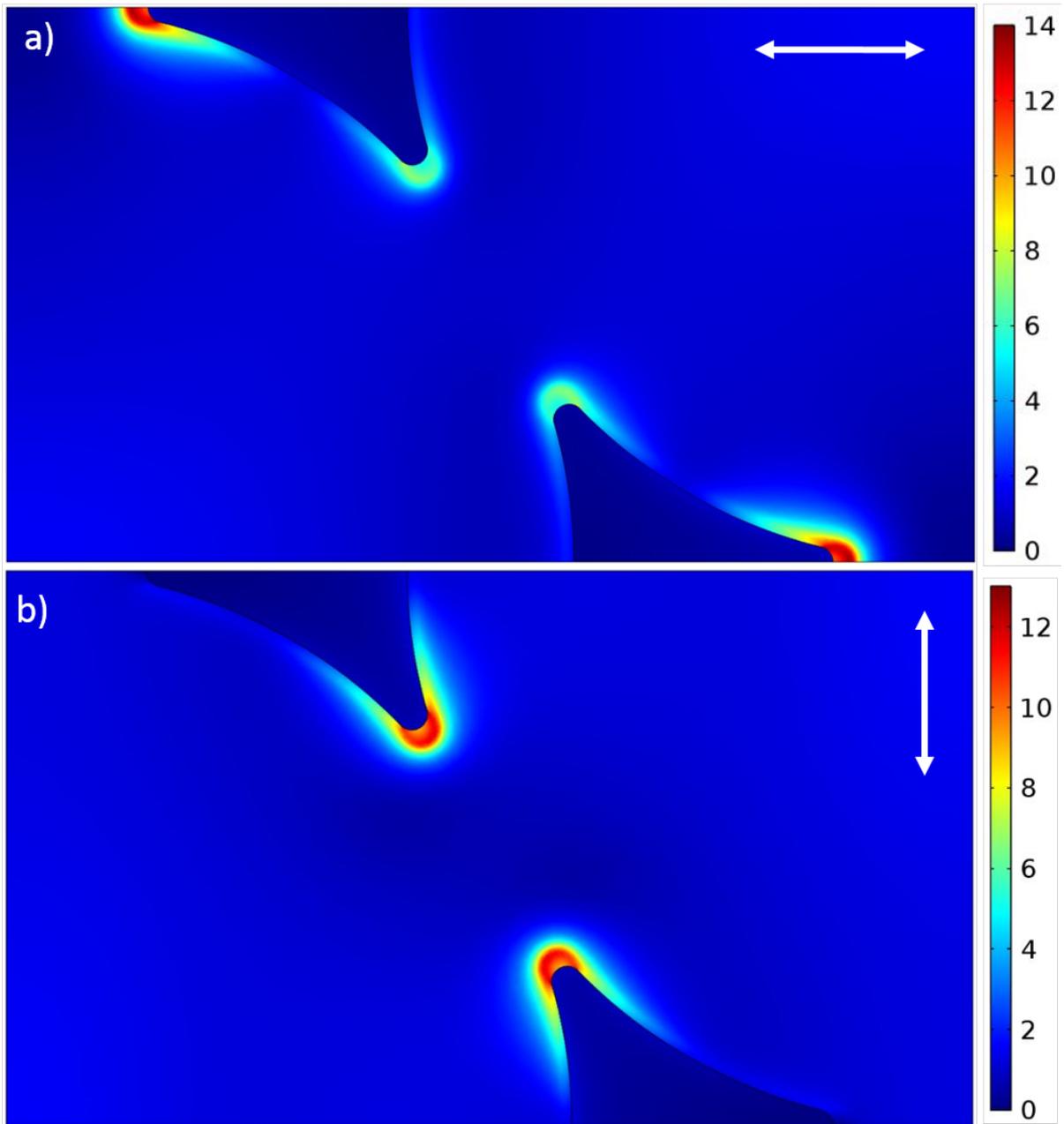


Fig. S14. Simulated fundamental dipole-like profile: the norm of the enhanced electric field (total field over incident field) is plotted 20 nm above the substrate at wavelength $2.55 \mu\text{m}$. (a) Horizontal incident polarization and (b) vertical polarization.