Supporting information to: Hybrid KTP-plasmonic nanostructures for enhanced nonlinear optics at the nanoscale

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1. Single crystal orientation determination by polarization analysis

In our work, single potassium titanyl phosphate (KTP) monocrystals have been studied with high numerical aperture objective for Second Harmonic Generation (SHG). This material has a significantly anisotropic second order nonlinear response tensor $\chi^{(2)}$, with the main component being $\chi^{(2)}_{z'z'z'}$. Under such tightly focused excitation beam, optical fields have to be considered as vectors and not only scalars, which makes any analytical study all the more complex.

We have then used a semi-analytical approach following[1,2] in order to find the Euler angles (ϑ, ϕ, ψ) of each crystal based on its polarization response in excitation. For a given discrete set of Euler angles, we have simulated the response of a crystal on a glass substrate in the laboratory frame, taking into account the whole $\chi^{(2)}$ tensor from the litterature.[3]

Then, for each experimental set of data $I_x(\theta), I_y(\theta)$ along x and y polarization directions, we compute the relative error between experimental and numerical data after normalization following:

$$\sum_{i=0}^{n}\left[\frac{\sqrt{\left(I_x(\theta_i)-I_{x,\mathrm{simu}}(\vartheta,\phi,\psi;\theta_i)\right)^2}}{\sum_{i=0}^{n} I_x(\theta_i)}+\frac{\sqrt{\left(I_y(\theta_i)-I_{y,\mathrm{simu}}(\vartheta,\phi,\psi;\theta_i)\right)^2}}{\sum_{i=0}^{n} I_y(\theta_i)}\right] \tag{1}$$

for each combination $(\vartheta,\phi,\psi)_{\mathrm{simu}}$ of Euler angles as defined on figure SS.1(a). From this, we get the best combination $(\vartheta,\phi,\psi)_{\mathrm{exp}}$ that minimizes this quantity, as can be seen on figure S.1(b) with the fit for (0°,0°,0°) at the top as compared with (20°,30°,20°) at the bottom. Figure S.1(c) shows the inverse of the error function of equation 1 as a function of angles ϑ and ϕ, where only the best result as a function of ψ is shown. This indicates the confidence level of the method.

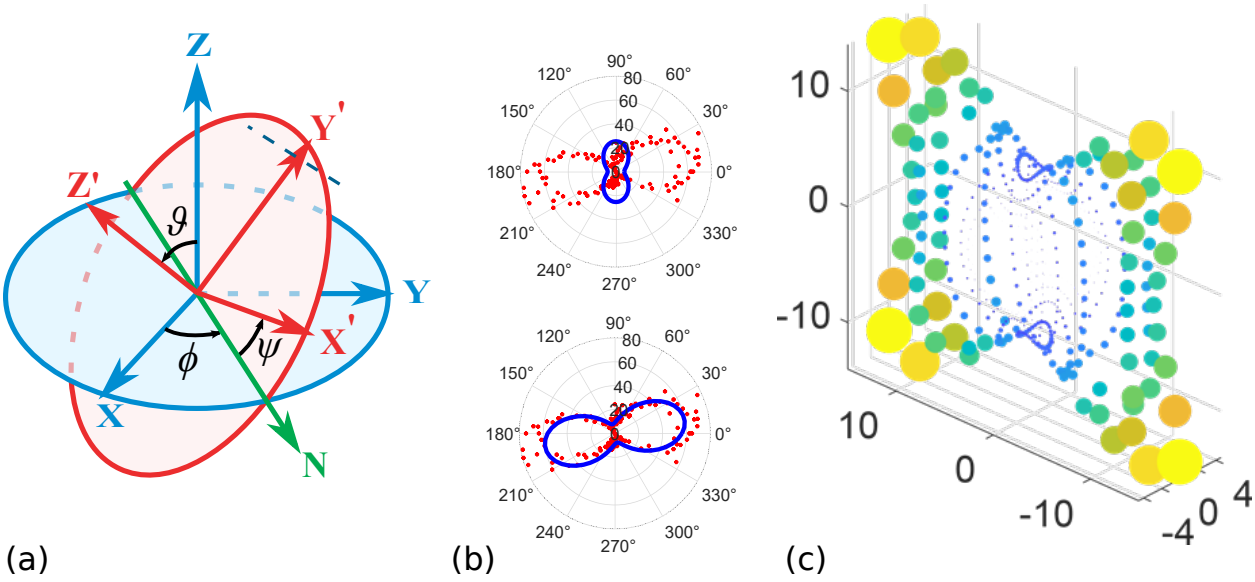

Figure S.1: (a) Definition of Euler angles (ϑ,ϕ,ψ) of the crystal, defined as the rotation between the laboratory reference frame (in blue) and the crystallographic frame (in red). (b) Experimental data and simulations respectively for (0°,0°,0°) (top) and (20°,30°,20°) (bottom) in the laboratory framework with respect to the excitation polarization angle. (c) As an example, the inverse of the relative error between simulations and an experimental data set for any combination of Euler angles is represented as a function of (ϑ,ϕ) in Cartesian coordinates: the bigger and the further from the origin the point is, the better the fit. Only the best fit as a function of ψ is indicated for each pair (ϑ,ϕ).

2. Comparison of simulated aluminum hybrid structures

We have run the same kind of analysis as in figure 5 of the main article, by replacing gold with aluminum, using reference[4] for the Rudnick and Stern parameter evaluation for this metal. As experiments for gold have been made with near-optimal antenna parameters, whereas aluminum structures were non resonant, we have run simulations with optimal antenna length (200 nm) based on the simulations presented on figure 4(b) of the main article. Results are shown on figure S.2, with a 84 nm wide and 25 nm thick crystal and 200x118x35 nm antennas separated by a 122 nm long gap centered on the crystal.

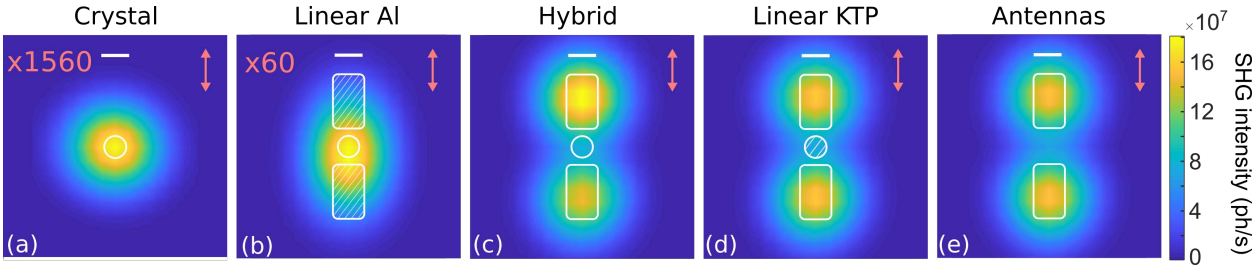

Figure S.2: Simulated excitation cartography of the SHG intensity for different nanostructures. (a) Single KTP crystal of 42 nm radius, 25 nm height and 10 nm minimum radius of curvature. (b), (c), (d) Hybrid Al-KTP structure respectively without the nonlinear contribution of the aluminum, with all contributions included and without KTP contribution. (e) Aluminum antennas without KTP. Color scale multiplying factors are indicated in red if applied.

Contrary to the case of gold, the nonlinear intensity of a hybrid structure remains almost unchanged when the contribution of the KTP crystal is removed, whereas it drops by a factor of 60 if the surface nonlinear contribution of aluminum is removed. Consequently, hypothesis **H2** does not apply here unlike gold.

References

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