Supplementary Information for

Nanostructured Amorphous Gallium Phosphide on Silica for Nonlinear and Ultrafast Nanophotonics

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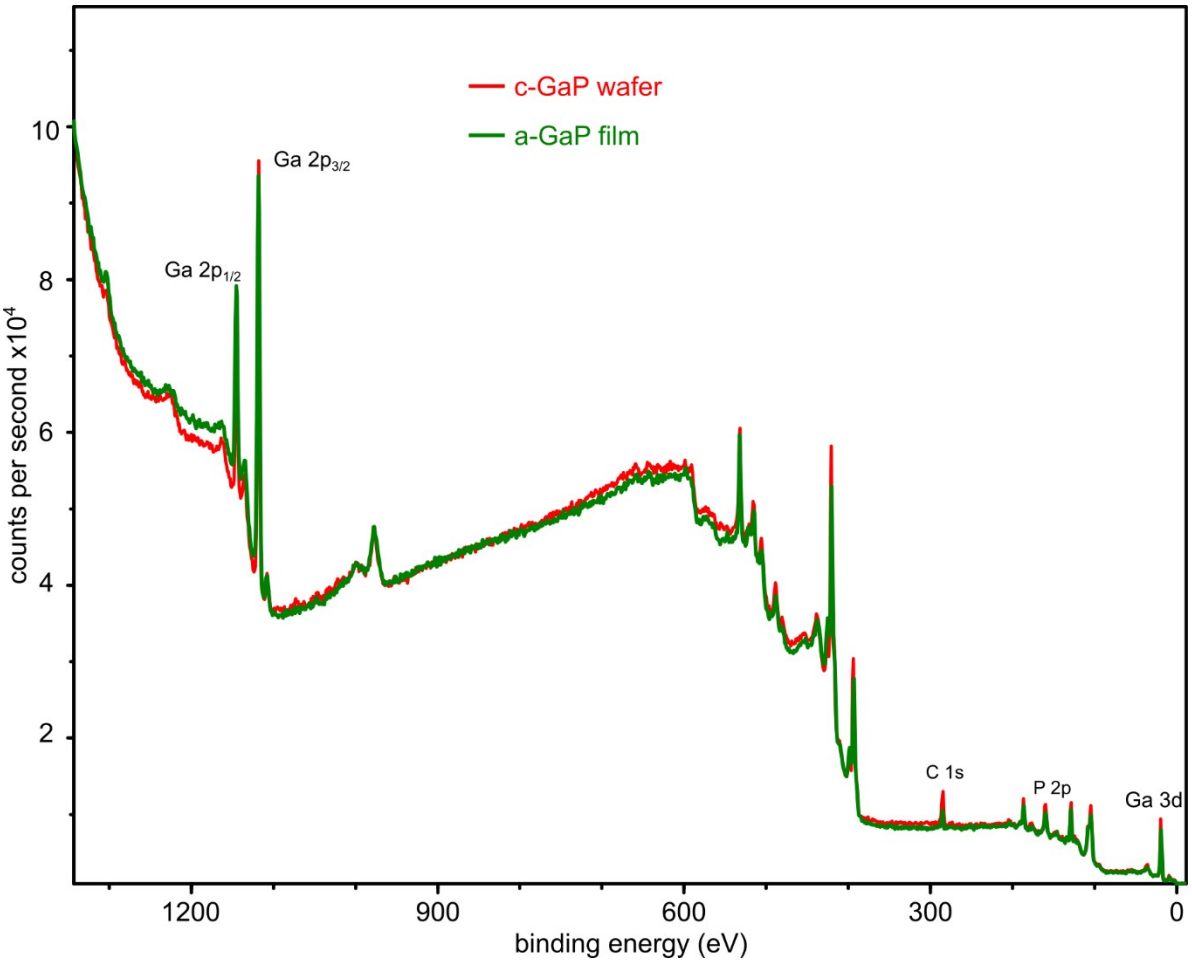

Figure S1: XPS measurement of the a-GaP film (green) and a c-GaP wafer (red) as reference sample with the important peaks indicated.

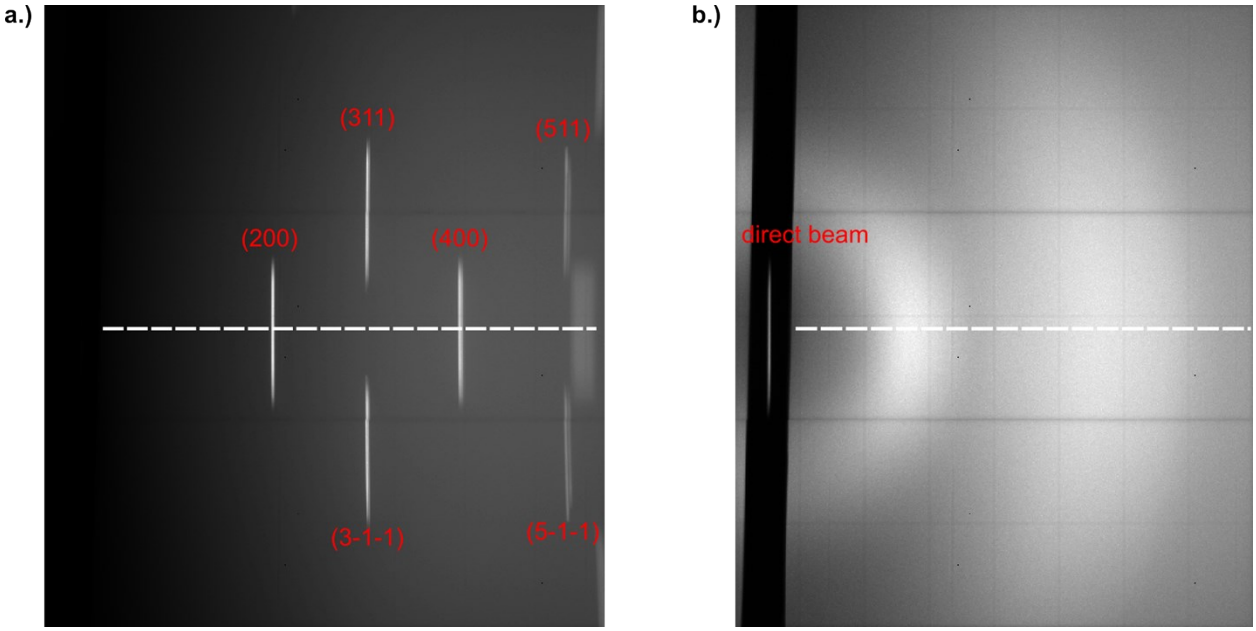

*Figure S2: Full XRD patterns of **(a)** the c-GaP wafer and **(b)** the a-GaP film. In the crystalline case, the peaks are marked with the corresponding Miller indices. The linear shapes result from the line-shaped X-ray beam. The dashed line indicates the position of the line-profiles that are shown in Figure 1a of the main text. The direct beam is weakened by an absorber casting a shadow on some are of the detector.*

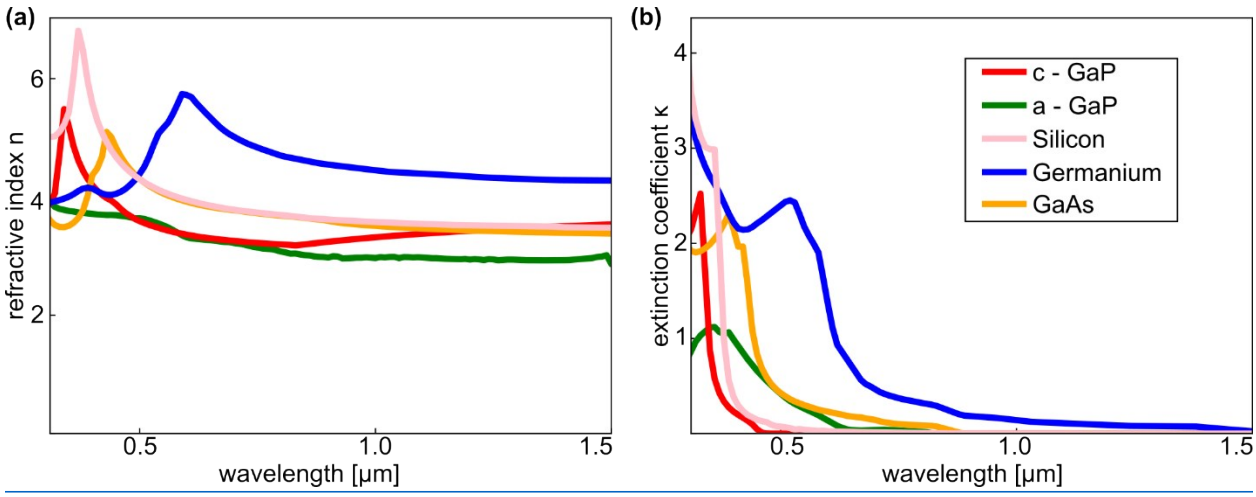

*Figure S3: **(a)** Refractive index n and **(b)** extinction coefficient κ for different semiconductors. For a-/c-GaP the data is identical to Figure 1b in the main text, for the other semiconductors the data were taken from the Lumerical FDTD material database which refers to **Handbook of Optical Constants of Solids I-III** by **E. Palik.***

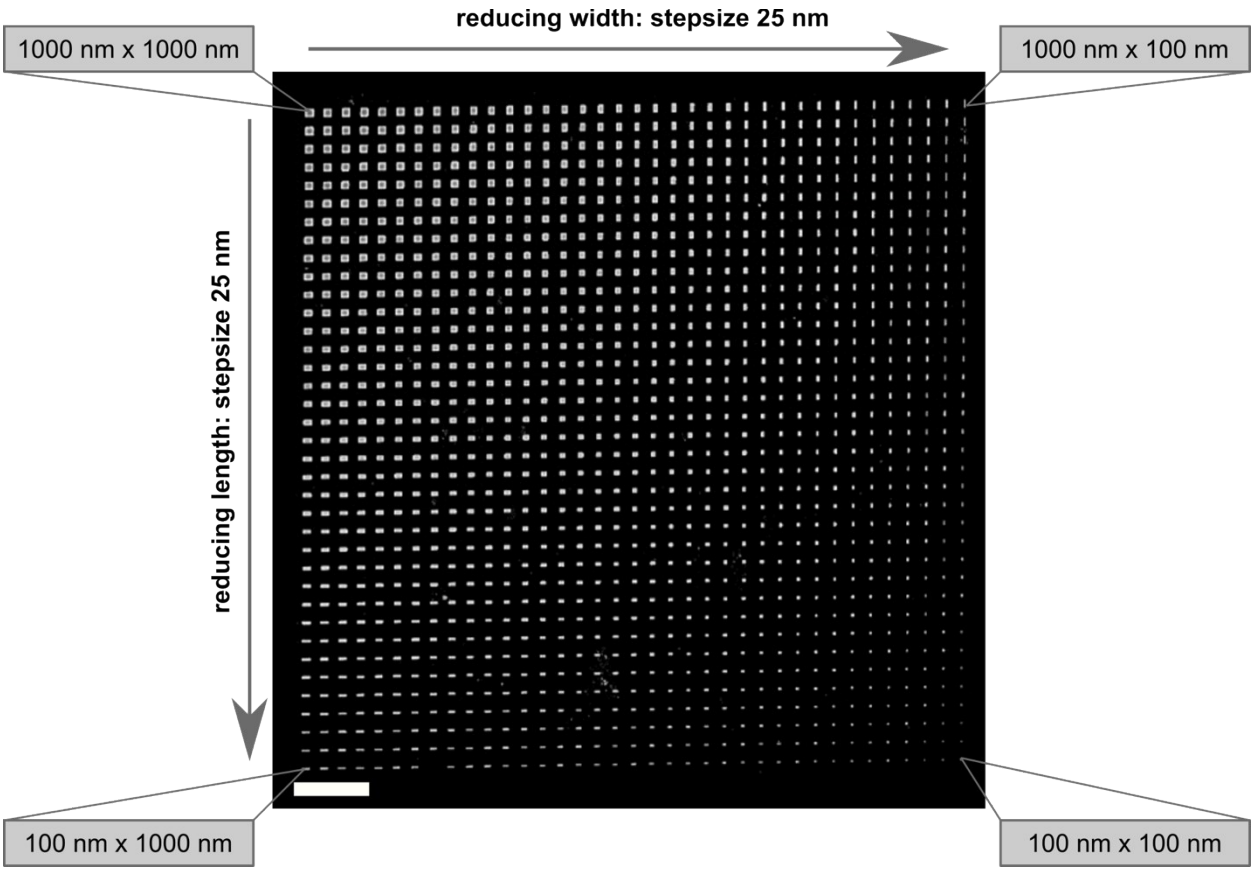

Figure S4: Details on the fabricated nanopatch array. The scale bar of the scanning electron microscope image corresponds to 10 µm.

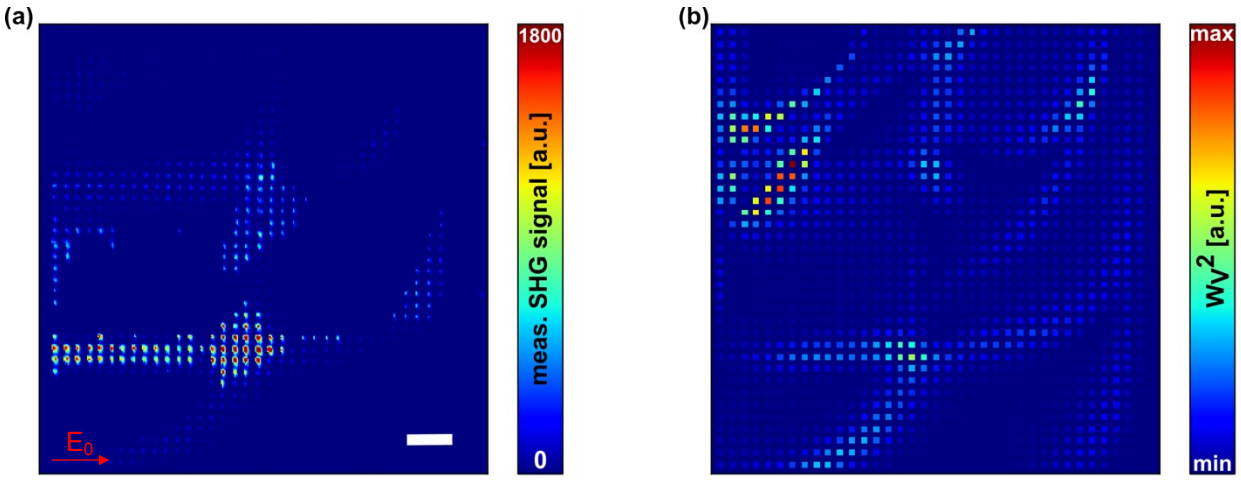

Figure S5: ***(a)*** *SHG measurement of the entire nanopatch array. The fundamental wavelength is* $1020\ nm$ *with an average power of* $50\ \mu W$*. Scale bar: 10* µm ***(b)*** *Numerical simulation of the square of the electric energy (W_V^2) for the array in (a).*

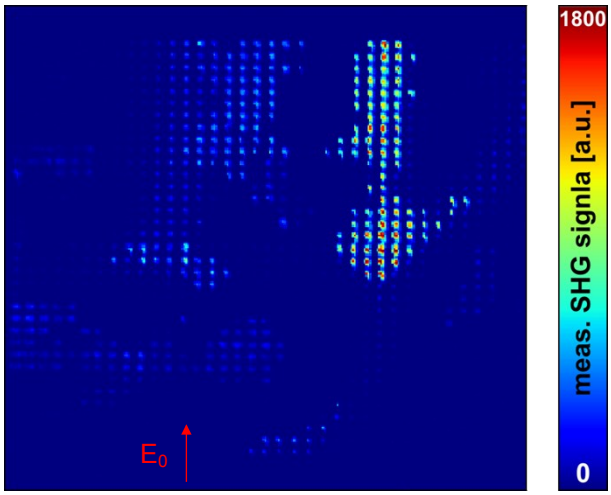

Figure S6: SHG measurement of the nanopatch array at 1020 nm and 50 µW excitation power (identical conditions as in the previous Figure S3), but with opposite (vertical) polarization.

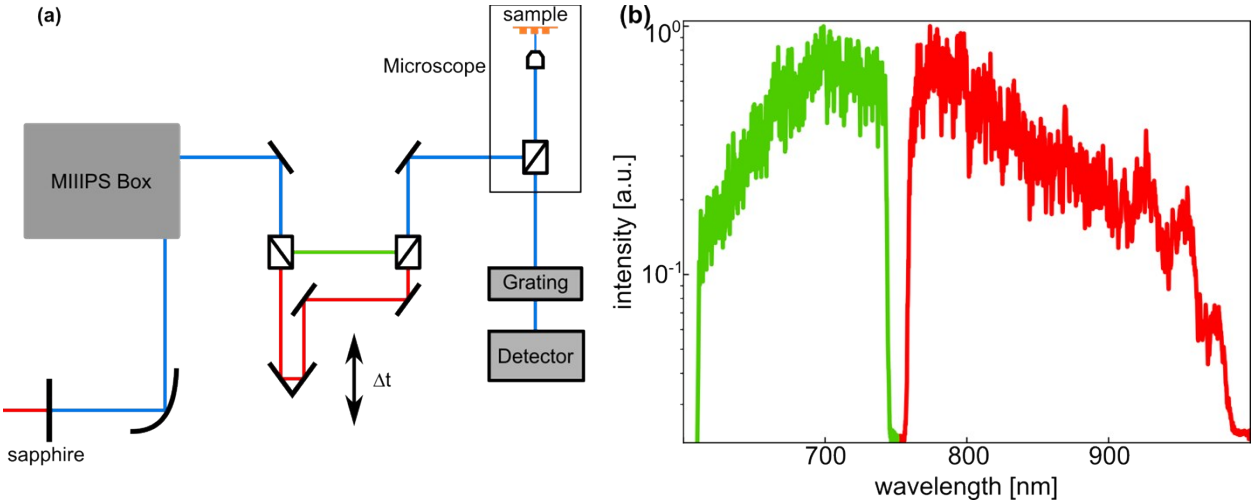

*Figure S7: **(a)** Experimental setup of the pump-probe experiment as described in Methods of the main text. **(b)** Measured spectrum of the supercontinuum light that is generated in the sapphire crystal, after the division with dichroic beam splitters. The colors represent the visible (green) and infrared (red) parts of the spectrum.*

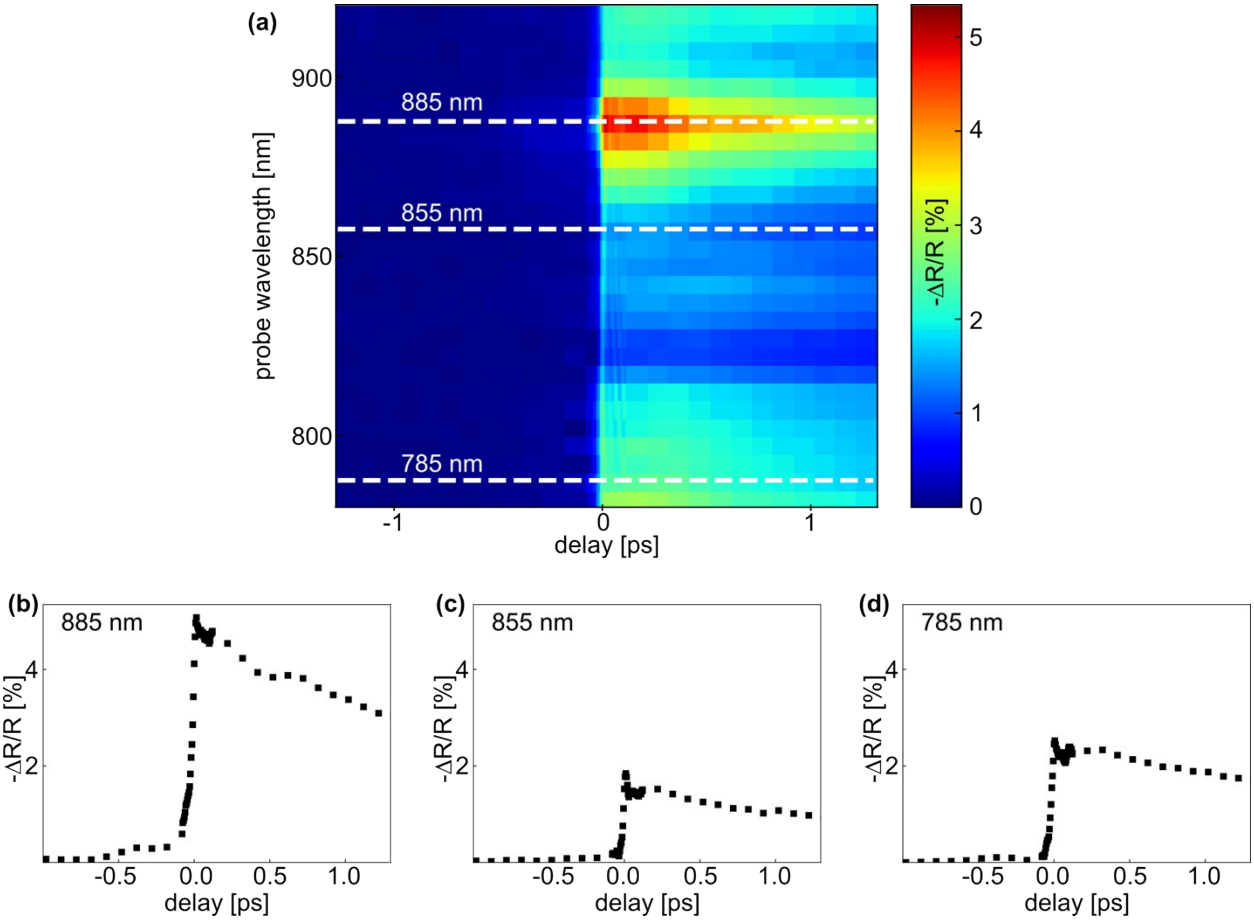

*Figure S8: Pump-Probe measurement of the square patch from the main text (Figure 4a) with the visible beam as pump and infrared beam as probe. **(a)** Spectral response of the differential reflectivity versus the pump-probe delay time. **(b)–(d)** Temporal traces at the indicated wavelengths, marked with the dashed lines in (a).*

	Im(ε) 500 nm	Im(ε) 850 nm	Ultrafast modulation	Free carrier response (after 1 ps)
a-GaP film*	3.1	0.0	< 1.75 %	< 0.4 %
a-GaP nanopatch*	-	-	< 5.6 %	< 1.8 %
c-GaP wafer[14]	0.0	0.0	< 70 %	0 %
c-GaP nanodisc[35]	-	-	< 40 %	0 %

*Table S1: Comparison of the ultrafast responses for crystalline and amorphous GaP, as-deposited and nanostructured. It should be noted that for the c-GaP wafer, the modulation of the transmittance ($\Delta T/T$) was measured, while the others consider the change in the reflectance ($\Delta R/R$). All values refer to the maximum reported values. * This work.*