Supplementary Material

Second-harmonic imaging of plasmonic Pancharatnam-Berry phase metasurfaces coupled to monolayers of WS_2

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1. Linear transmission spectra of the metasurface samples

The plasmonic resonances influence the light-matter interaction and therefore the nonlinear frequency conversion. For our work, we assume that only the plasmonic resonance at the fundamental wavelength will influence the nonlinear SHG process. Therefore, we measured the linear transmission spectra of the samples to analyze the spectral mode position. Furthermore, we want to ensure that there are no resonances of the plasmonic nanorods in the visible regime, which can lead to additional coupling effects and modification of the SHG emission efficiency. Figure S1 shows the experimentally obtained and numerically calculated transmission spectra for the metasurface samples with the three different antenna lengths. Note that the experimentally obtained transmission spectra for the NIR spectral region are shown in Figure 1B of the main text.

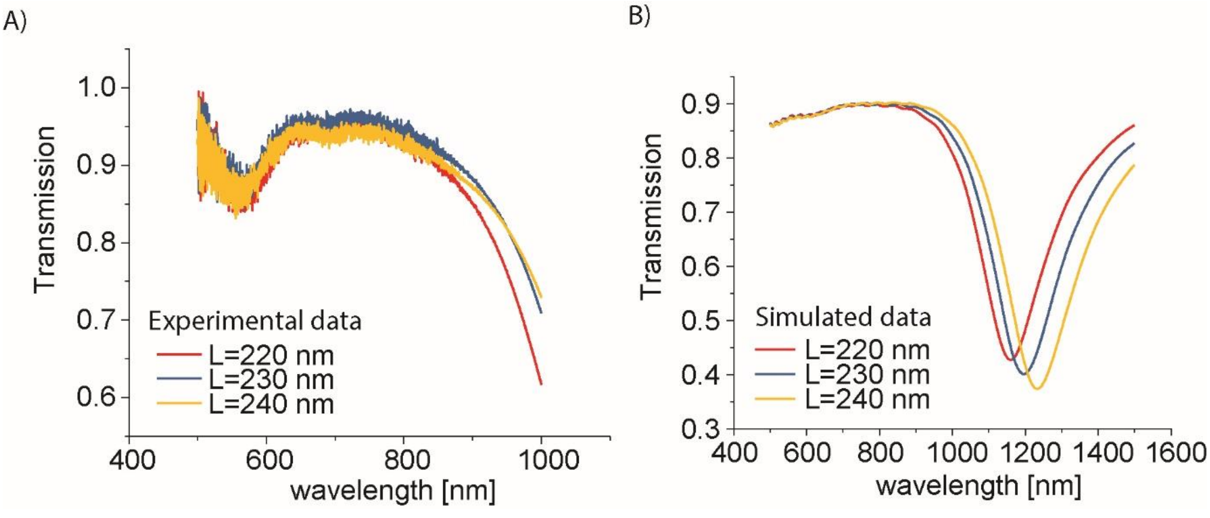

Figure S1: Linear transmission spectra for the hybrid metasurfaces. (A) Linear transmission spectra of the plasmonic nanorod metasurfaces with the length of 220-240 nm in the visible regime. The resonance dip occurring in the near-infrared region, starting at around 900 nm, shows the onset of the plasmonic resonance at around 1200 nm. The measurement was taken with unpolarized light (B) Simulated transmission of the periodic nanorod array predicting a strong plasmonic resonance around 1200 nm. The simulations are confirmed by the experimental results shown in Figure 1 of the main text.

In the visible region, the transmission spectra of the metasurfaces show only a small resonance dip at around 550 nm. This resonance is sufficiently spaced from the SHG wavelength at around 640 nm, where the spectrum is nearly flat. Hence, we expect no modification of the SHG emission due to a resonance of the nanorods in the visible range. Furthermore, the simulated transmission spectra used for the design of the nanorod length as shown in Figure S1B are well confirmed by the experimental spectra (Figure 1B).

2. Power dependency

A crucial parameter is the power of the illuminating laser beam. Since the nonlinear response in the co-polarization state σ^+ of the hybrid metasurface is lower than the response in the cross-polarization state σ^-, we performed a power-dependent measurement of the nonlinear signal. Such measurement provides also information about depletion or damaging effects that can occur due to the finite carrier concentration in the WS_2 or the heating effects by the absorption of the gold nanoantennas. For a weak perturbation regime, we expect a power-dependent SHG signal following the relation $I_{\mathrm{SHG}} \propto P^2$ for the incident fundamental beam power P. Figure S2 illustrates the power-dependent SHG measurement of the sample.

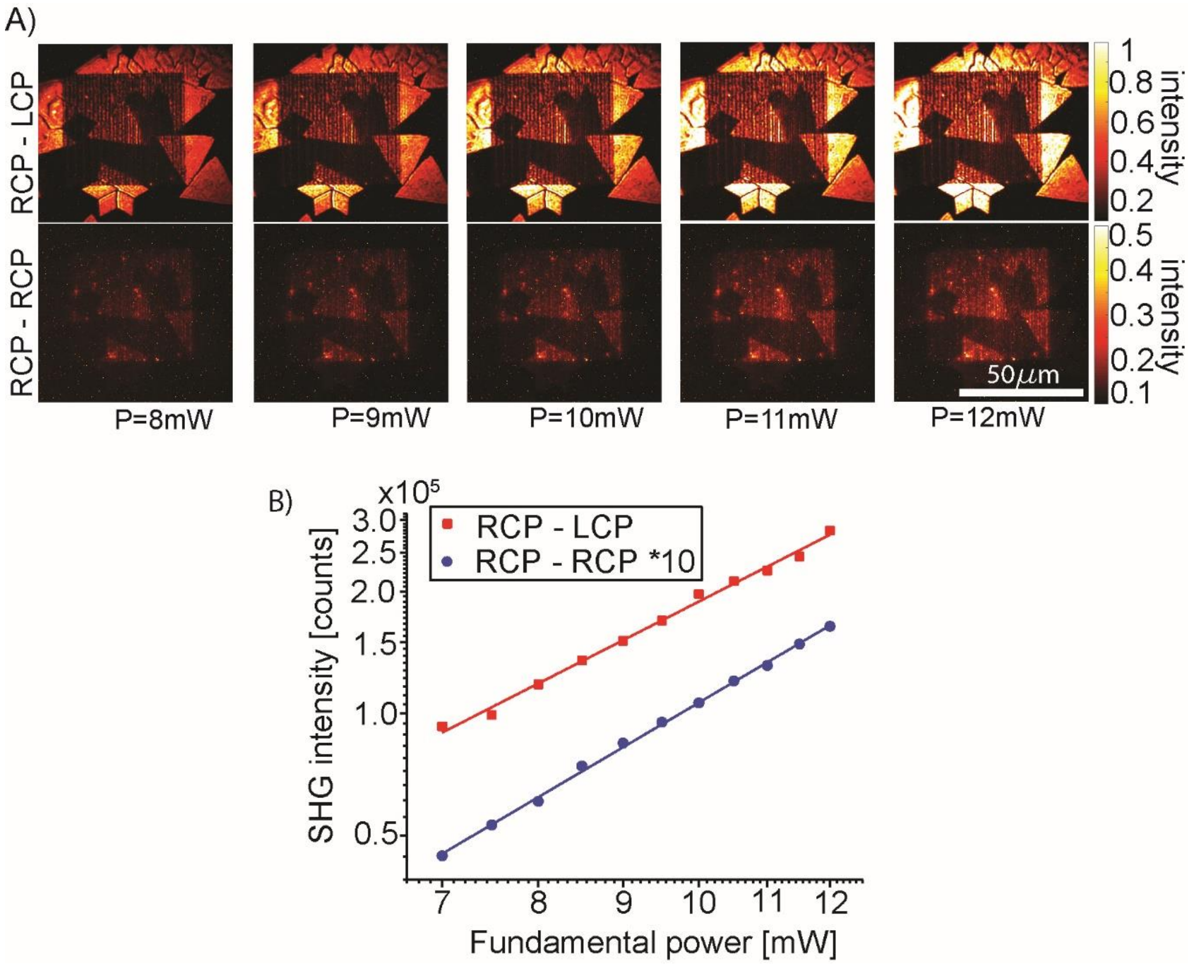

Figure S2: Power dependent measurement of the SHG signals. (A) Spatially resolved SHG signal from the hybrid metasurface and the 1L-WS_2 flakes in cross-polarization state σ^- and co-polarization state σ^+ for different incident fundamental powers. (B) Extracted SHG intensities from the images in (A) versus the incident power for both polarization states. Note that the values for the SHG signal for the co-polarization state (blue dots) have been multiplied by the factor 10. The solid lines represent a polynomial fit to the measured values with the slopes of 2.08 for the fit of RCP-LCP and 2.41 for the fit of RCP-RCP.

By increasing the power of the incident laser beam, the real-space images of the hybrid metasurface in cross- and co-polarization state σ^- and σ^+ brighter, indicating a stronger SHG signal. By plotting the integrated intensity versus the incident power, we obtain a nearly quadratic increase of the SHG signal (Figure S2B). By fitting the measured data with a polynomial function, we obtain a slope of 2.08 for the cross-polarization state σ^- and a slope of 2.41 for the co-polarization state σ^+. As in the ideal case for a second harmonic generated signal, both of them should be ~2 to confirm a quadratic relation. The calculated slopes are in the same regime; therefore, a quadratic relation can be assumed.

Furthermore, the SHG signal does not show any saturation effects for the higher power values which leads us to the conclusion that we did not exceed the damage threshold of the samples. While the real-space images of the cross-polarization state σ^- are visible at all powers, for the co-polarization state σ^- we require at least ~10 mW power to measure the signal with sufficient signal-to-noise ratio.

3. Phase information - cross-polarization state σ^-

We now investigate the dependency of the phase information implemented in the plasmonic metasurface on top of 1L-WS_2 on the polarization state. After a fundamental beam with no encoded phase information interacts with the hybrid metasurface, two SHG beams are scattered, one in each polarization state (σ^+ and σ^-). As already shown, the SHG beam in the co-polarization state σ^+ carries the phase information, while the SHG signal in the cross-polarization state σ^- is not phase-modulated at all. To verify this assumption, we investigate a hybrid metasurface with encoded phase information in the plasmonic metasurface for the cross-polarization state σ^-.

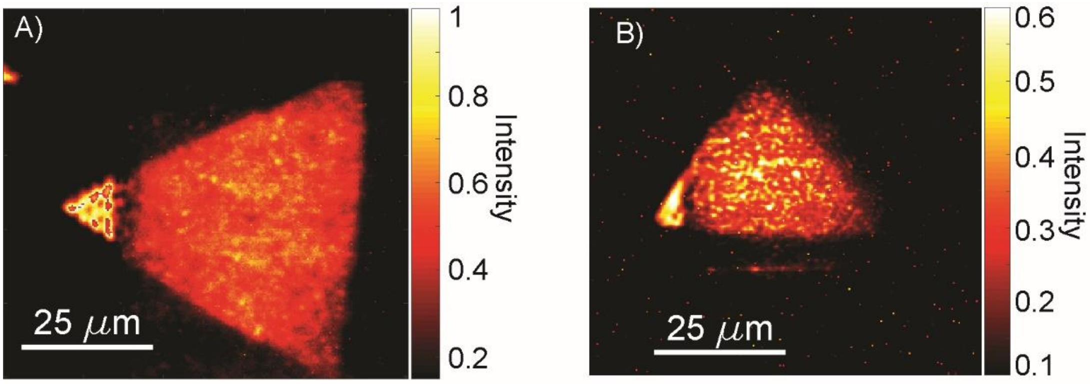

Figure S3: SHG measurement of the cross-polarization state σ^- (A) SHG image of the same flake as shown in Figure 4B but measured in the cross-polarization state σ^-. This polarization state does not carry the PB phase information and therefore no interference effect is observable. (B) SHG image of the same flake as in Figure 4C also measured in the cross-polarization state σ^-. This polarization state does not carry any phase information and therefore no spatial interference effect is observable.

Figure S3 illustrates the SHG images of the same hybrid metasurfaces shown in Figure 5B-C in the main text but measured in the cross-polarization state σ^-. Both images do not show any spatially modulated SHG intensity for the hybrid area. This is in correspondence with the PB phase which is only carried by the σ^- state of the fundamental light scattered by the nanorods and therefore appears only in the σ^+ state of the SHG signal.

4. SHG intensity ratios

To fortify the working principle of the hybrid metasurface, we calculate the intensity ratios of the SHG between the pure WS_2 and the hybrid metasurface case. Figure 3E shows two areas with both cases for co- and cross-polarization state. The area 1 correspond to the hybrid metasurface case, while area 2 marks the WS_2 with covered by the metasurface. We determine the SHG signal strength originating from area 1 corresponding to the metasurface case with SHG_1 and regarding to the non-metasurface SHG_2, respectively.

The SHG intensity ratio for the cross-polarization σ^- state is calculated to $\frac{\mathrm{SHG}_1^{\sigma^-}}{\mathrm{SHG}_2^{\sigma^-}} = 0.54$ with respect to the sizes of both areas. The comparison of the two areas shows, that the hybrid metasurface area (area 1) emits about 46% less SHG signal in the cross-polarization state than the bare 1L-WS_2 area. Contrary to the lower SHG signal in the cross-polarization state, a strong SHG signal in the usually forbidden co-polarization state for the hybrid metasurface is observed. The image for the co-polarization state σ^+ shows a dark area 2, where no SHG light is emitted for the non-metasurface case. A more meaningful statement is the ratio between the SHG signal in area 2 in the cross-polarization state ($\mathrm{SHG}_2^{\sigma^-}$) and the signal in area 1 in the co-polarization state ($\mathrm{SHG}_1^{\sigma^+}$). This ratio indicates the percentage of SHG light emitted from the co-polarization state by the hybrid metasurface and is calculated to $\frac{\mathrm{SHG}_1^{\sigma^+}}{\mathrm{SHG}_2^{\sigma^-}} = 0.15$.