Supporting Information for

Giant Enhancement of Four-Wave Mixing by Doubly Zone-Folded Nonlocal Metasurfaces

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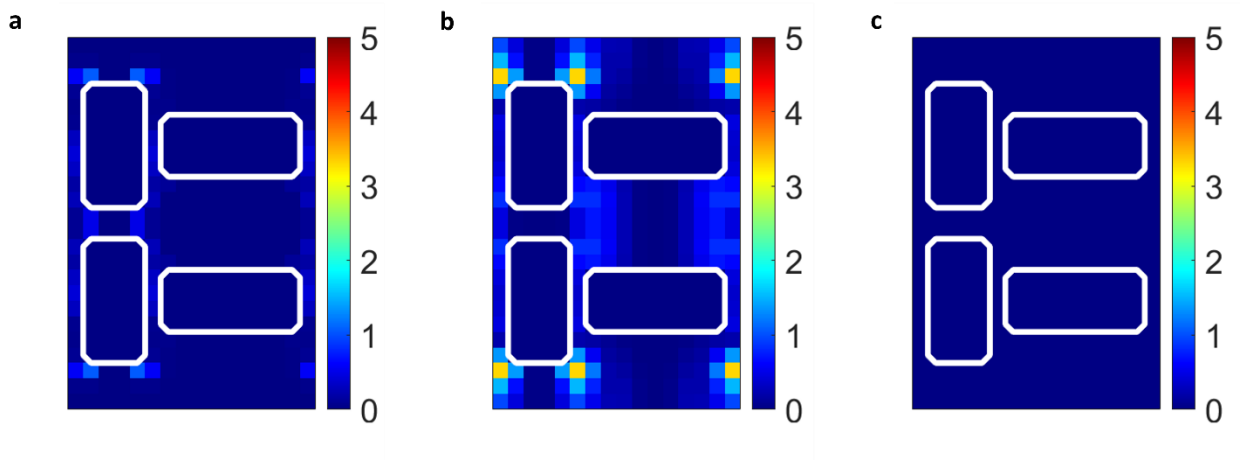

Figure S1: Calculated field overlap figure of merit $|E(\omega_1)E(\omega_2)^*E(\omega_1)|$ for design in Figure 2 of Main Text for field components with minimal contribution to enhancing four-wave mixing. (a) E_Y excited by *x*-polarized incident light (b) E_Z excited by *x*-polarized light (c) E_X excited by *y*-polarized light.

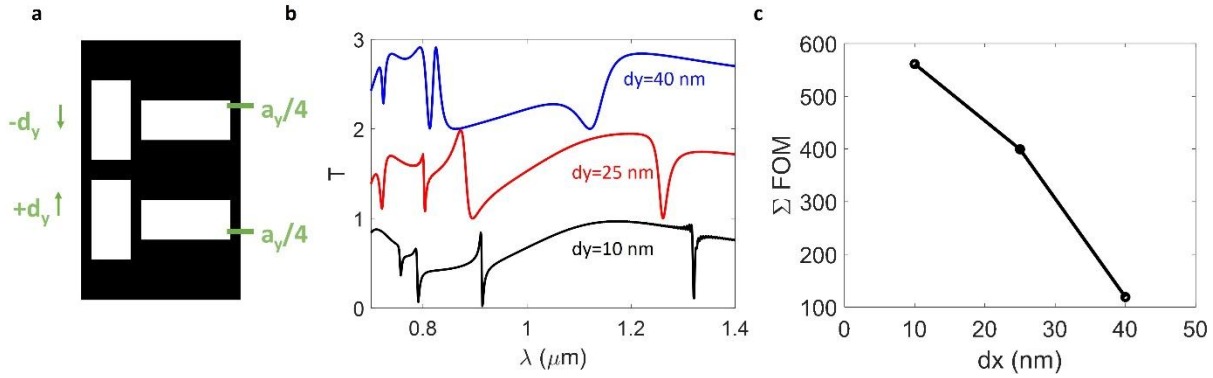

Figure S2: Selection of Q-factor and field overlap through control of *y*-direction dimerizing perturbation. (a) Definition of magnitude of *y*-direction dimerizing perturbation d_y (d_y=25 nm in example schematic). (b) Simulated transmission spectra for three different values of d_y. (c) Calculated sum of field overlap figure of merit $FOM = |E(\omega_1)E(\omega_2)^*E(\omega_1)|$ in the silicon for three different values of d_y.

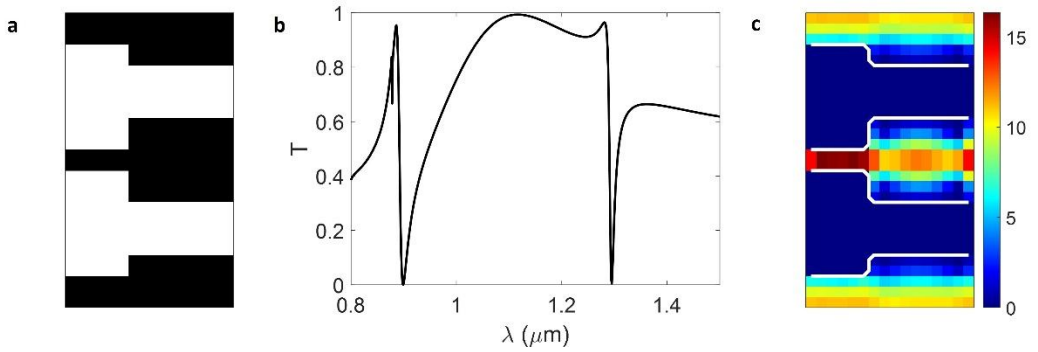

Figure S3: Simulation of metasurface in Fig. 4 accounting for fabrication imperfections that joins adjacent nanovoids in *x*-direction. (a) Simulated spectra for *x*-polarized incident light. (b) Calculated field overlap figure of merit $|E(\omega_1)E(\omega_2)^*E(\omega_1)|$.

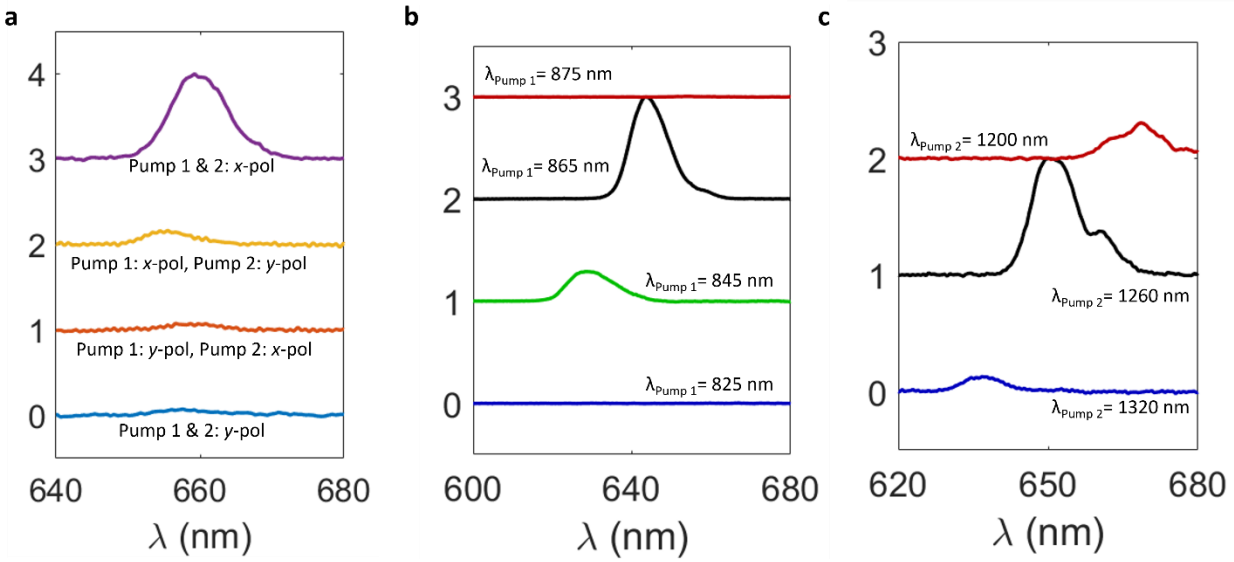

Figure S4: Additional nonlinear spectral measurements of device in Fig. 5. (a) Expanded spectral measurements from Fig. 4c of main text at 0 ps time delay including measurements where only one pump is resonant with its respective q-BIC (*x*-polarized). (b) Spectral measurements with varied Pump 1 wavelength and $\lambda_{Pump\ 2}$=1290 nm. Both pumps are *x*-polarized and 2 mW average power. (c) Spectral measurements with varied Pump 1 wavelength and $\lambda_{Pump\ 1}$=865 nm. Both pumps are *x*-polarized and 2 mW average power. All measurements normalized to maximum signal in plot and offset by 1 for visual clarity.

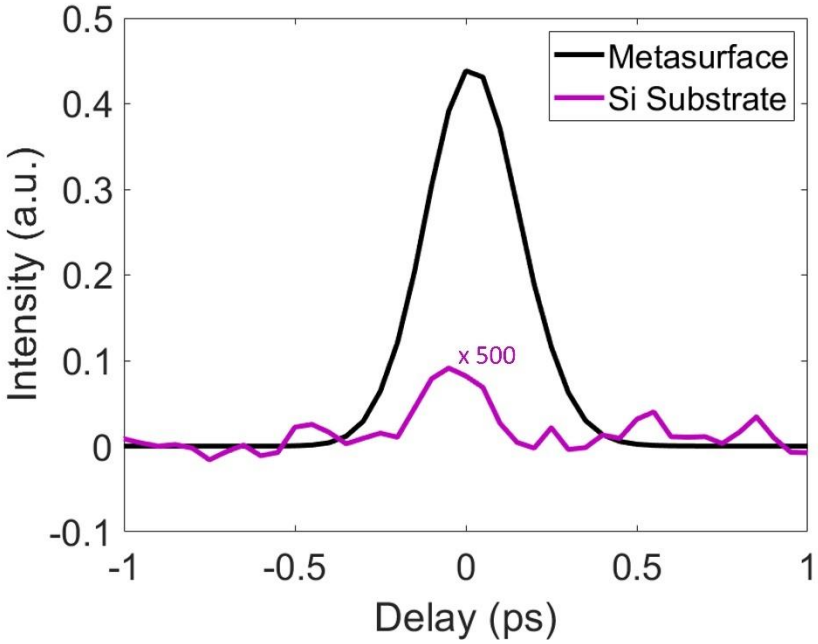

Figure S5: Additional confirmation of ~2400x enhancement of FWM from device in Fig. 4 of the main text. Measured intensity as a function of time delay between pump beams on metasurface (Black curve) and unpatterned si-on-glass substrate (purple curve, magnified by a factor of 500 for visibility) measured at a different spot on the metasurface than Fig. 4. Power of both pumps is 10 mW.

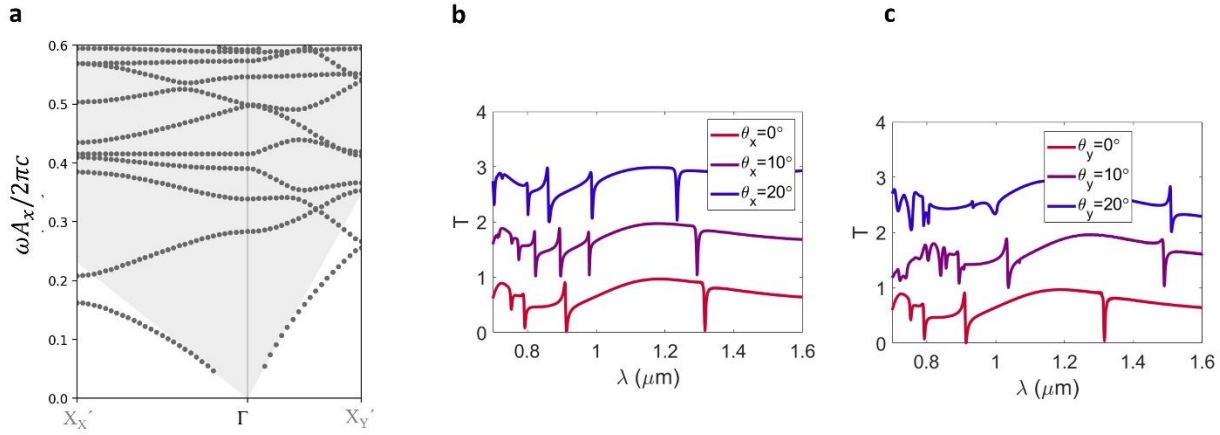

Figure S6: Calculated angular dispersion of device in Fig. 4 of the Main Text. (a) Calculated band structure. (b-c) Simulated transmission spectra as a function of incident angle for light that is off-normal in the *x*- direction (b) and *y*- direction (c).

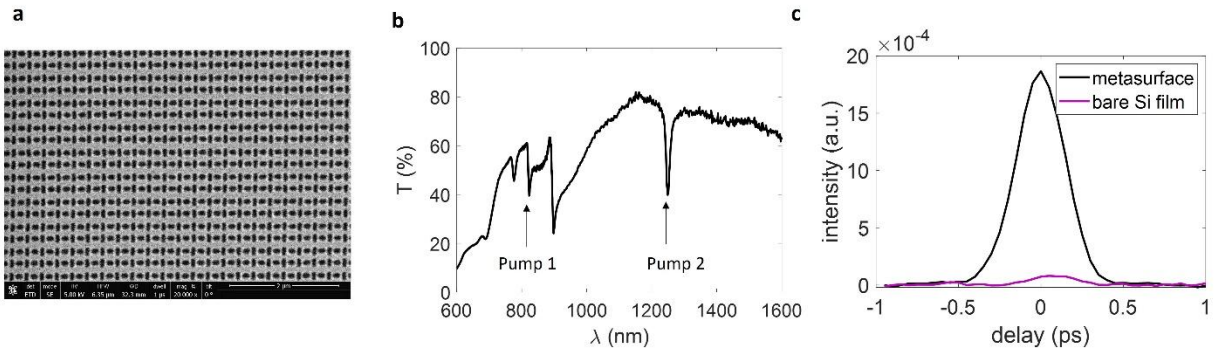

Figure S7: Measured device with improved fabrication and a higher Q-factor for Pump 2 (Q_{Pump1}~110, Q_{Pump2}~ 170) but only ~23x enhancement of four-wave mixing when pumped on resonance. (a) Scanning electron micrograph. (b) Measured linear spectrum for *y*-polarized incident light. (c) Measured intensity as a function of time delay between pump beams on metasurface (Black curve) and unpatterned Si-on-glass substrate (purple curve). Power of both pumps is 2 mW.

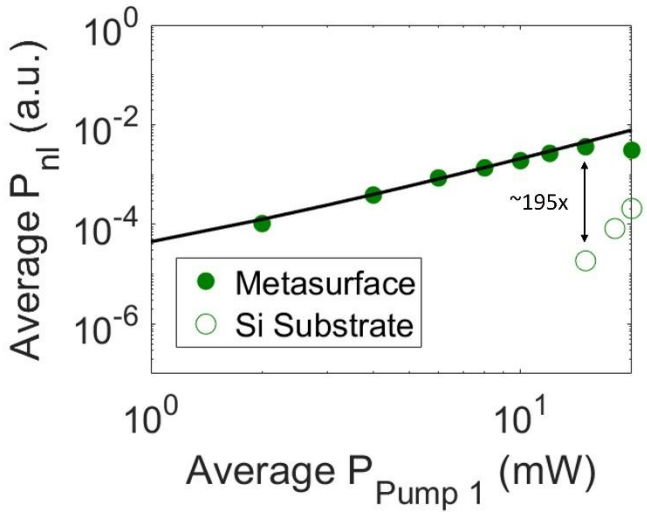

Figure S8: Nonlinear signal as a function of Pump 1 of the device in Figure 4 of main text. Pump 2 is *y*-polarized and 2 mW.

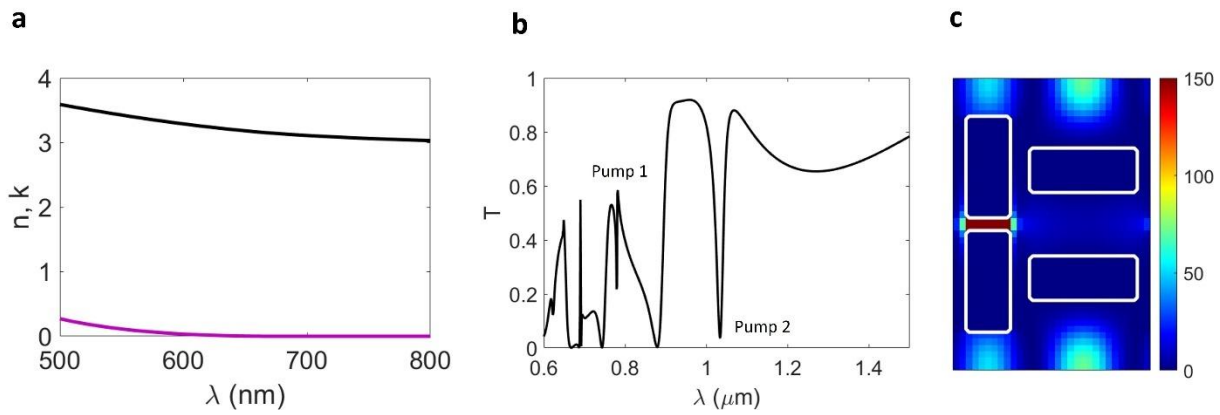

Figure S9: Design of silicon rich nitride metasurface in Figure 5 of Main Text. (a) Measured optical constants. (b) Simulated spectrum of metasurface for *x*-polarized incident light with lattice constants a_x=300 nm and a_x=450 nm and *y*-direction spacing perturbation d_y=24 nm. (c) Calculated $E_x(\omega_1)\, E_x(\omega_2)^*\, E_x(\omega_1)$ in quadromer lattice (with apertures etched into the thin film outlined in white) as a figure of merit for enhancement of four-wave mixing following nonlinear polarization.

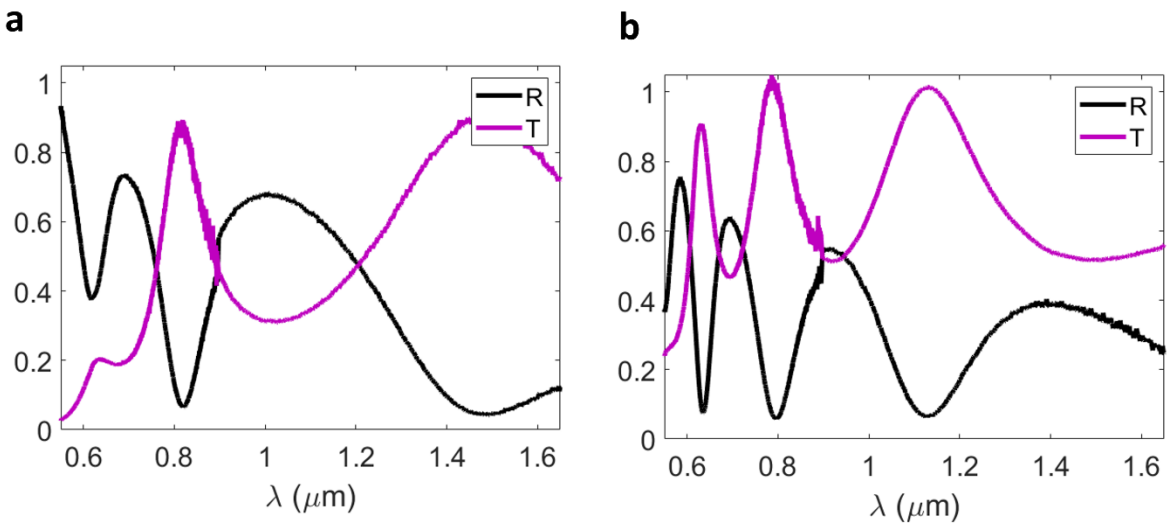

Figure S10: Optical characterization of unpatterned ~200 nm Si-on-glass and ~440 nm SRN-on-glass substrates used in Fig. 4 and 5 of the Main Text. (a-b) Linear reflection and transmission spectra for (a) silicon and (b) SRN.