

## **Tailoring the ultrafast and nonlinear photonics of MXenes through elemental replacement**

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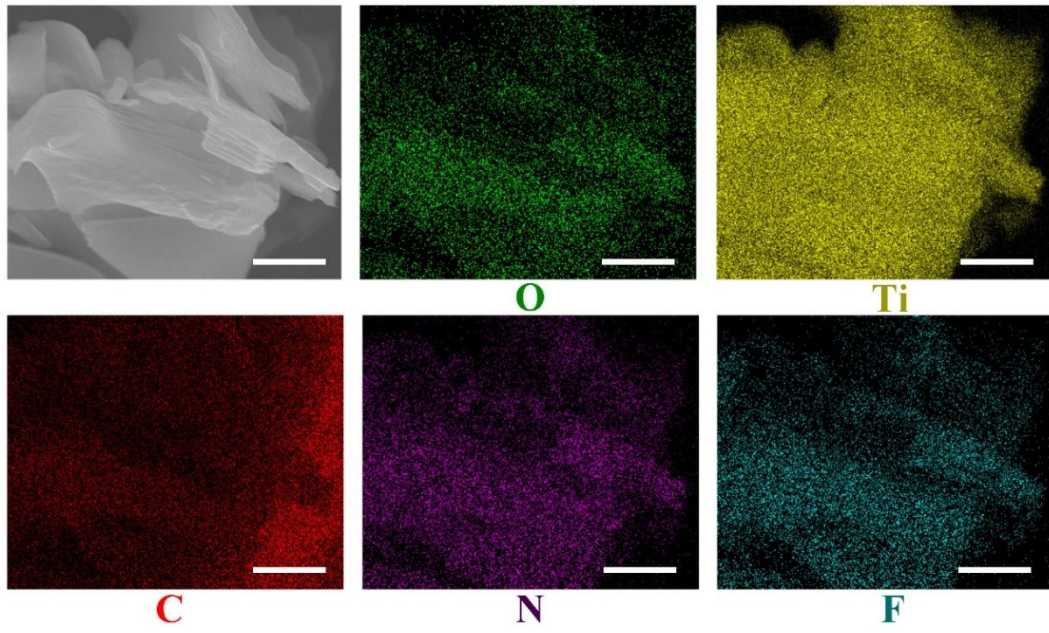
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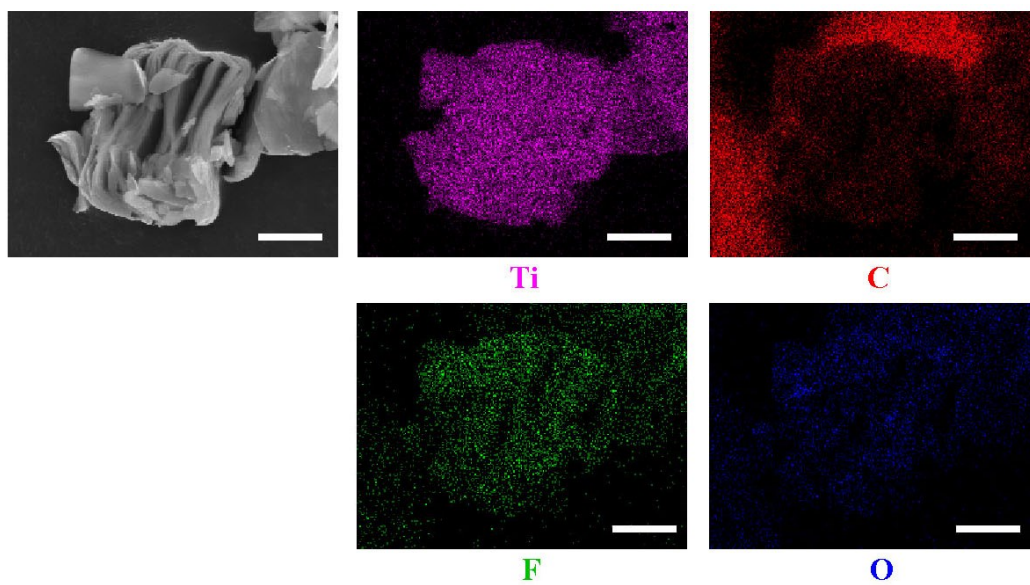
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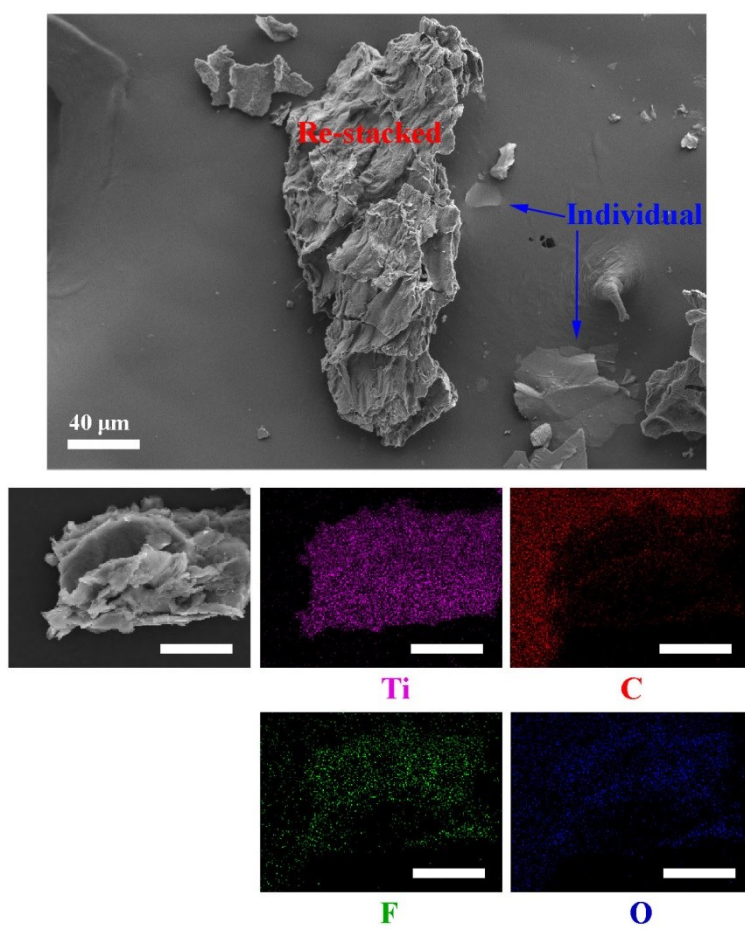
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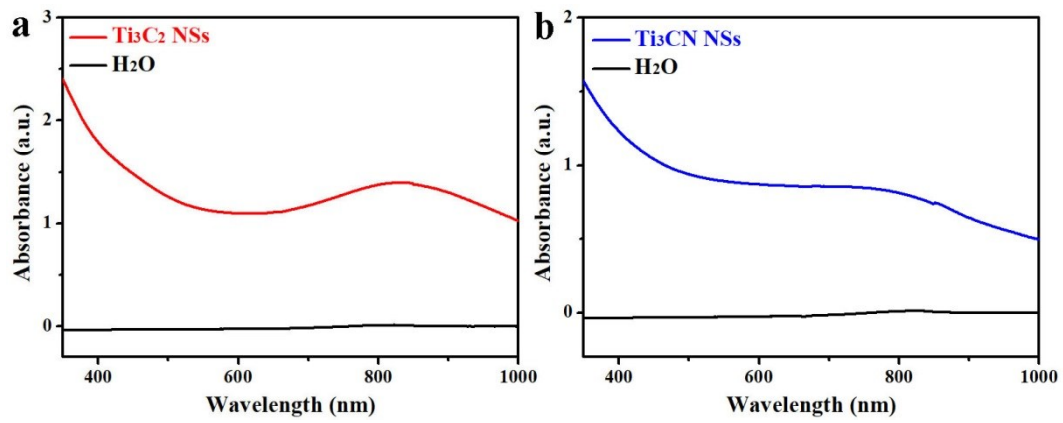
**Figure S1.** The EDS mapping of multi-layer  $Ti_3CN$ . Bar is 2.5  $\mu m$ .



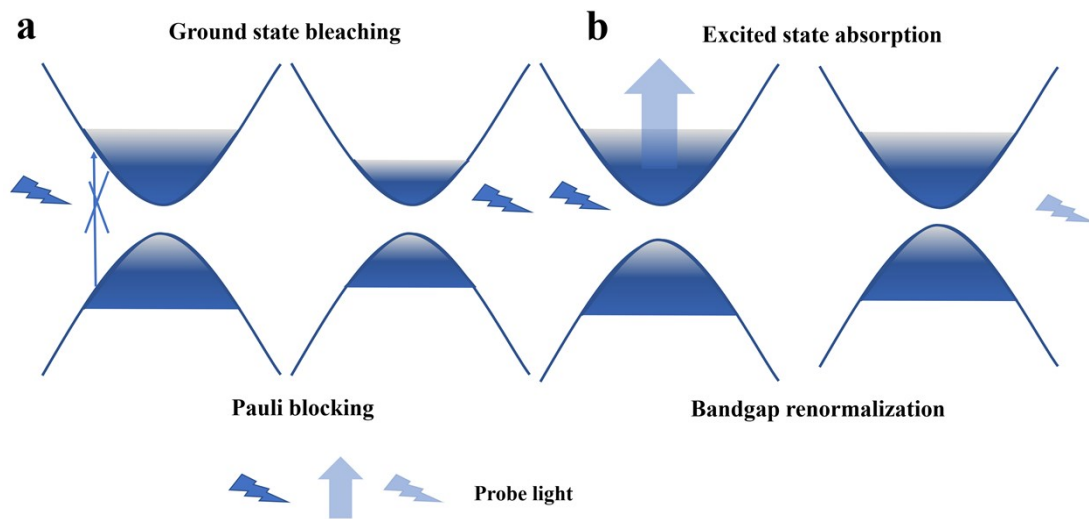
**Figure S2.** The EDS mapping of multi-layer  $\text{Ti}_3\text{C}_2$ . Bar is 5  $\mu\text{m}$ .



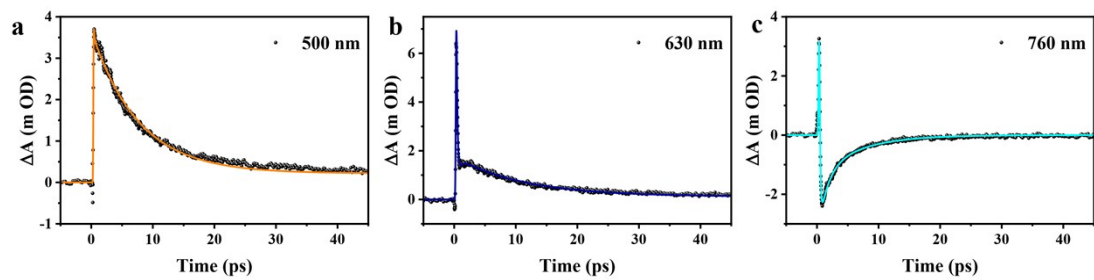
**Figure S3.** SEM image of delaminated  $\text{Ti}_3\text{C}_2$  flakes and the corresponding EDS mapping image. Bar is 10  $\mu\text{m}$ .



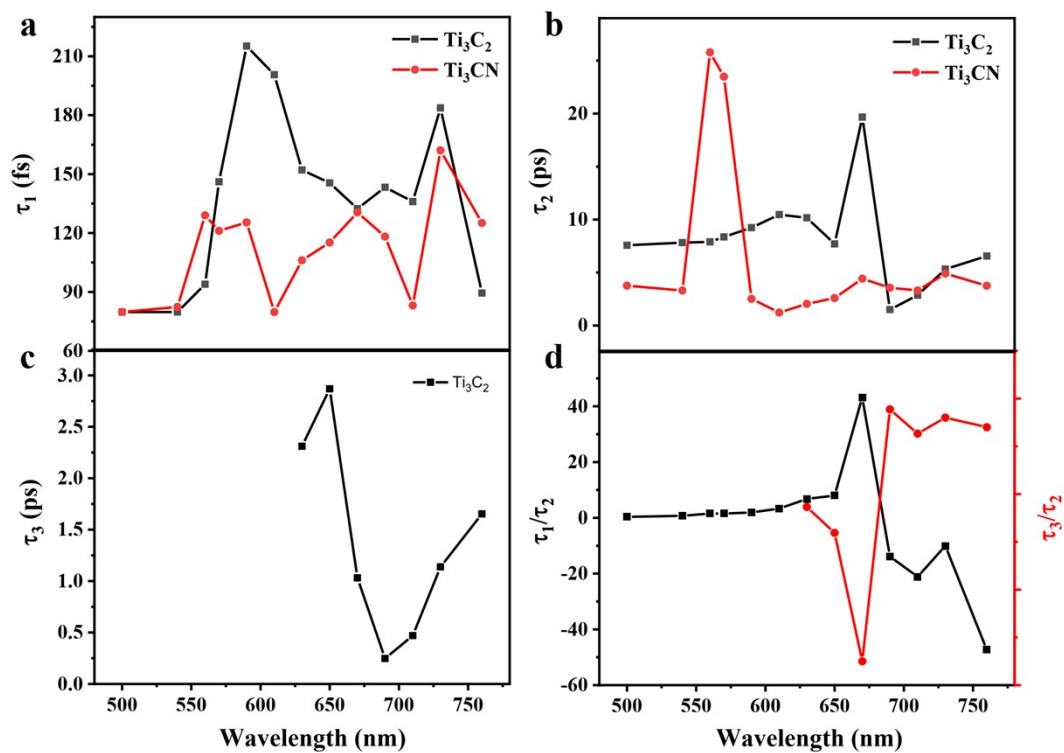
**Figure S4.** UV-Vis spectra of few-layer Ti<sub>3</sub>C<sub>2</sub> (a) and Ti<sub>3</sub>CN (b) NSs in water.



**Figure S5.** The schematic illustration of ground state bleaching and excited state absorption.

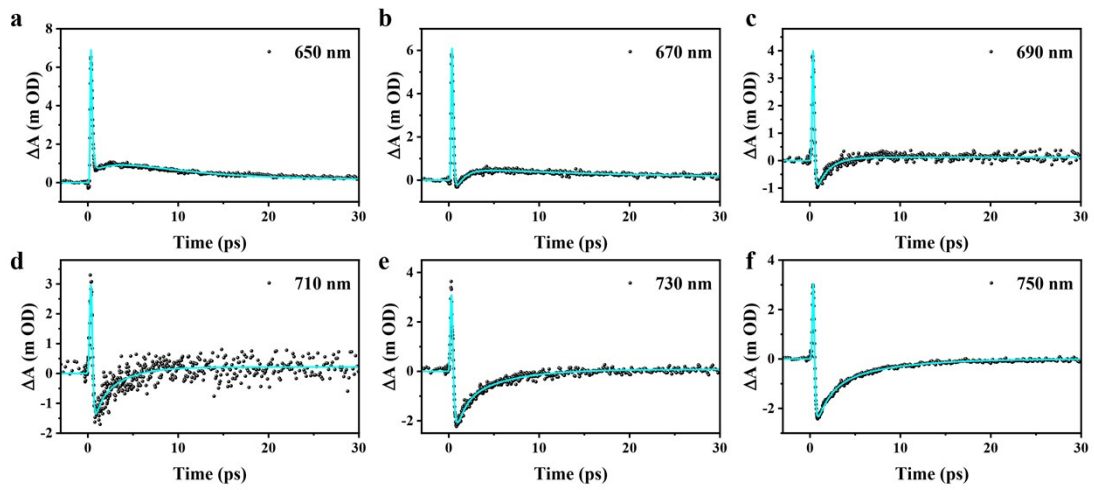


**Figure S6.** Dynamic curves in MXene NSs. (a-c) The experimental data and fitted results of the  $\text{Ti}_3\text{C}_2$  at 500 nm, 630 nm, 760 nm, respectively.

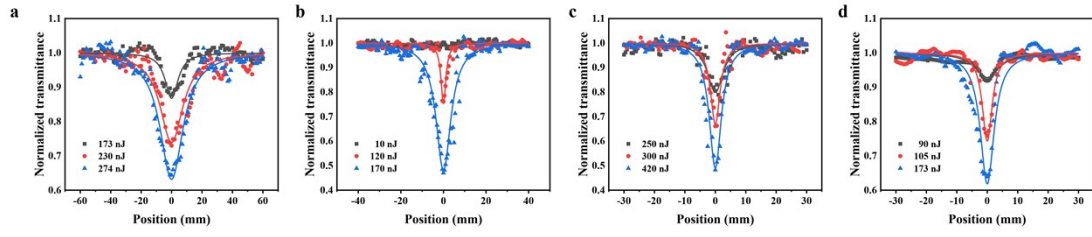


**Figure S7.** The summaries of fitted time constants at different probe wavelengths. (a) Carrier-carrier scattering time components, (b) carrier-phonon scattering time components of two MXenes, (c) BGR time components of  $\text{Ti}_3\text{C}_2$  NSs, (d) the comparison of fitted time components in  $\text{Ti}_3\text{C}_2$  NSs.

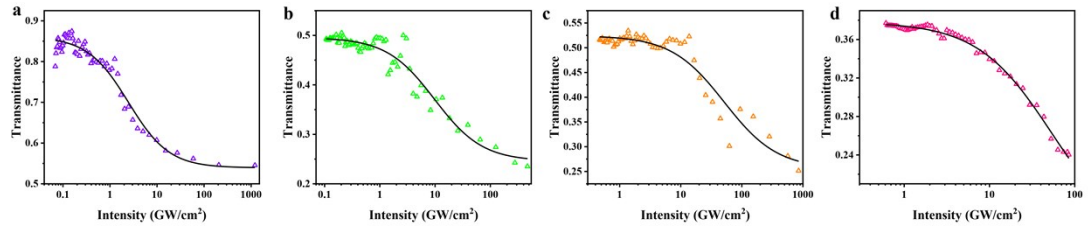




**Figure S8.** The BGR components dynamic curves in  $\text{Ti}_3\text{C}_2$  NSs under different probe wavelengths of (a) 650 nm, (b) 670 nm, (c) 690 nm, (d) 710 nm, (e) 730 nm, and (f) 750 nm.



**Figure S9.** OA Z-scan traces and fitted curves of MXenes NSs. (a)-(d) NLO response of Ti<sub>3</sub>C<sub>2</sub> NSs at 500 nm, 600 nm, 700 nm and 800 nm with different incident intensities.



**Figure S10.** The nonlinear absorption characteristics of MXene NSs. (a)-(d) NLO response of Ti3CN NSs under various wavelengths with maximum incident single pulse energy.