

Supporting Information

Structurally Defined Nano-scale Sheets from Self-Assembly of Collagen-Mimetic Peptides.

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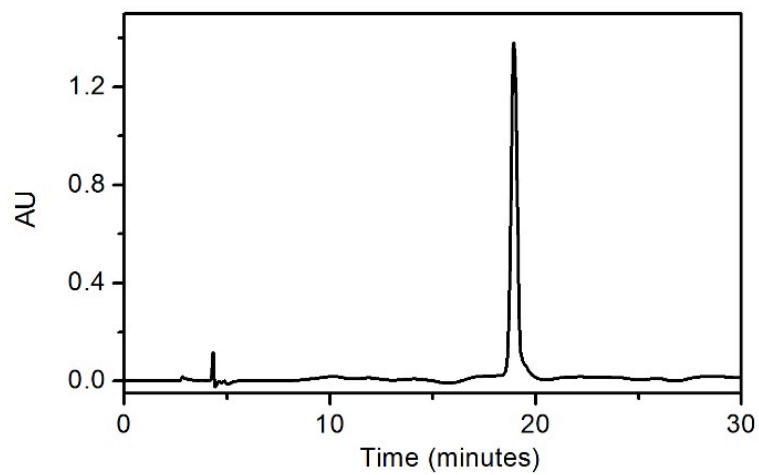


Figure S1. Analytical HPLC trace of a purified sample of peptide **NSI**.

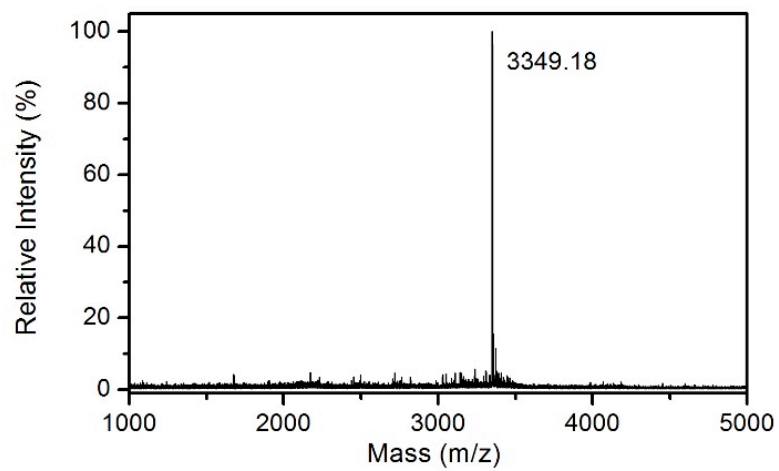


Figure S2. MALDI-TOF mass spectrum of peptide **NSI** after HPLC purification.

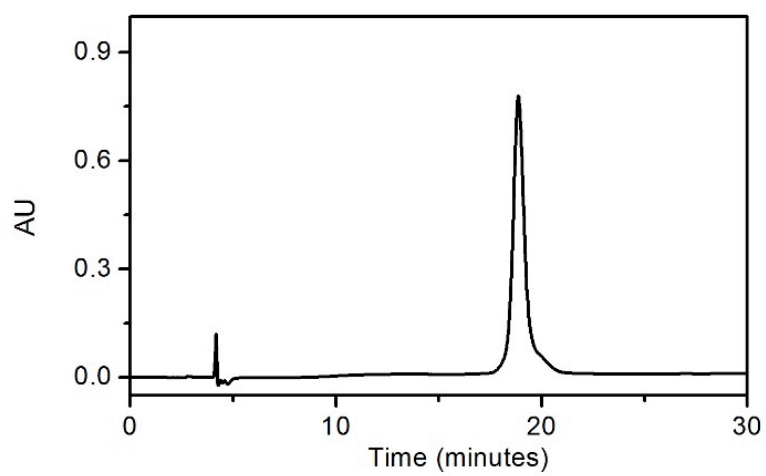


Figure S3. Analytical HPLC trace of a purified sample of peptide **NSII**.

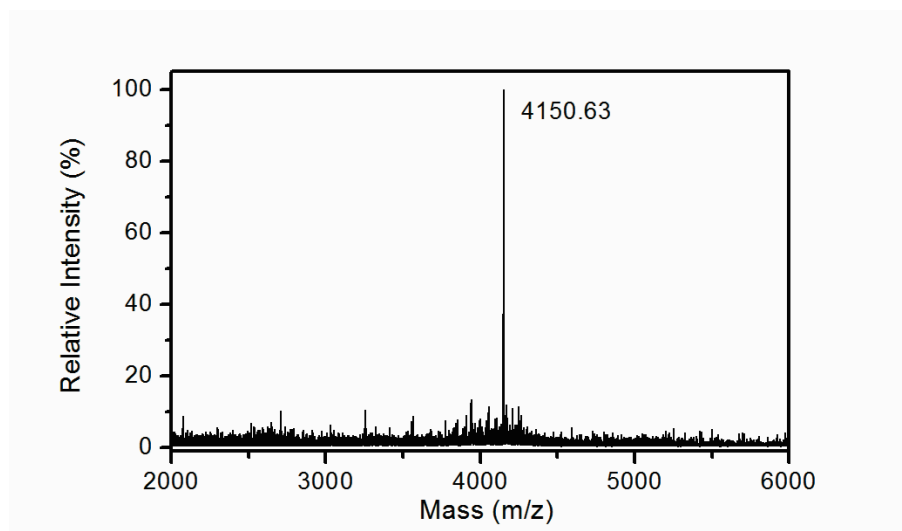


Figure S4. MALDI-TOF mass spectrum of peptide **NSII** after HPLC purification.

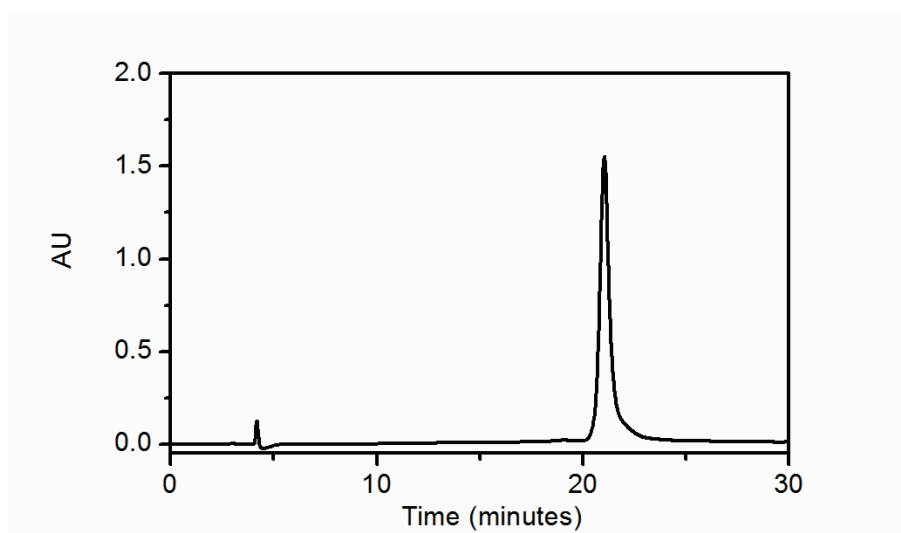


Figure S5. Analytical HPLC trace of a purified sample of peptide **NSII***.

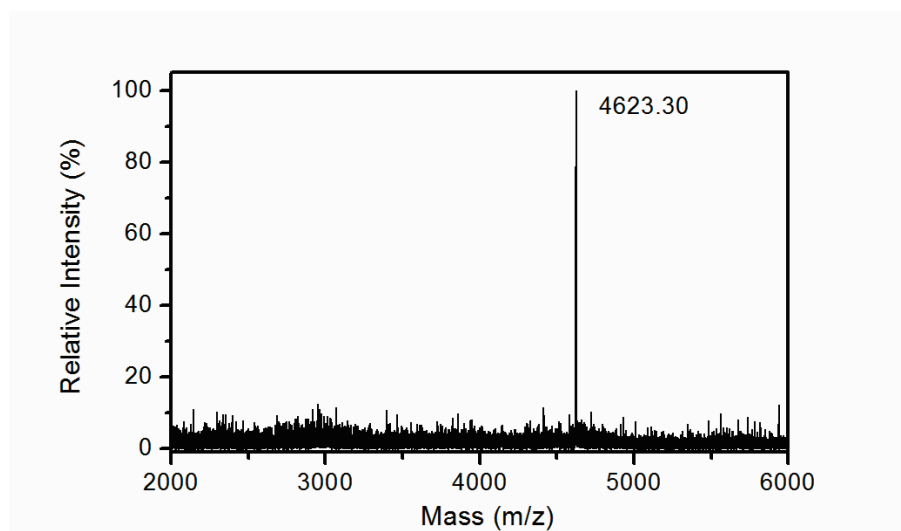


Figure S6. MALDI-TOF mass spectrum of peptide **NSII*** after HPLC purification.

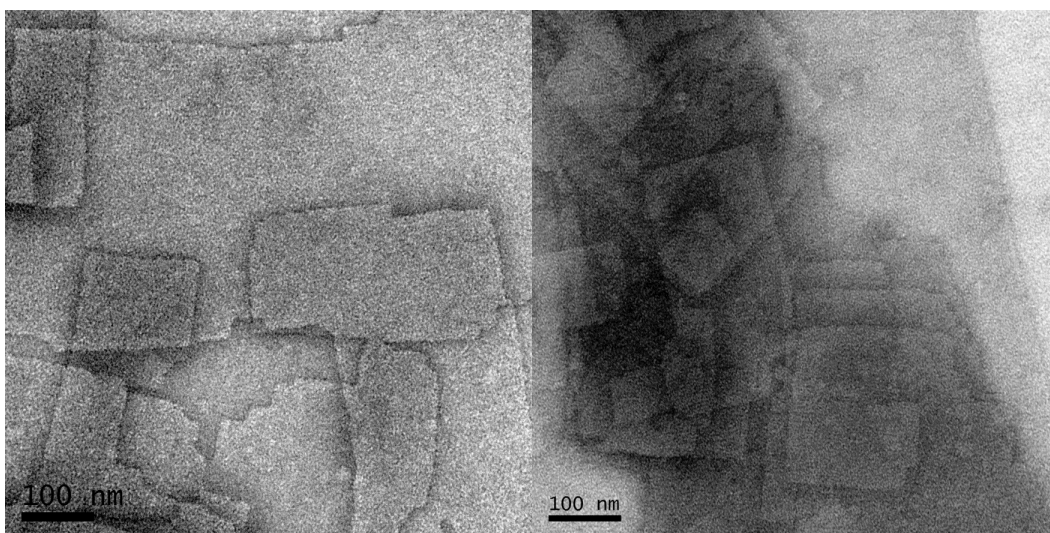


Figure S7. Sheet-like assemblies of peptide **CP11** (2 mg/mL) in MOPS buffer (20 mM, pH 7.0). Note the irregular edges of the assemblies.

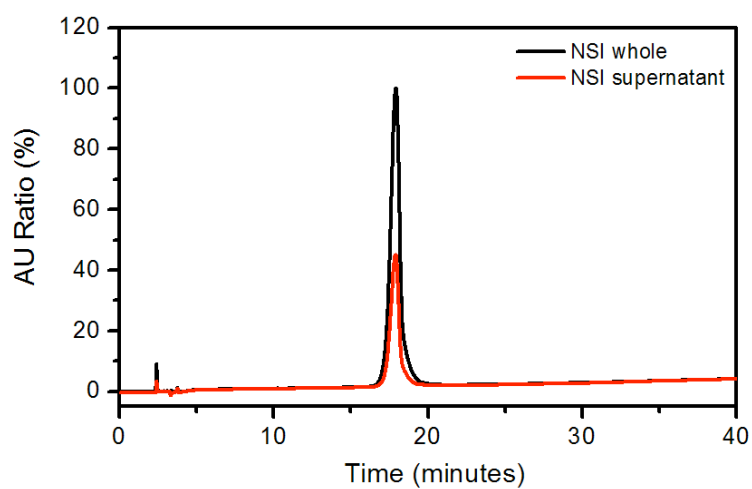


Figure S8. Analytical HPLC analysis of **NSI** (2 mg/mL) in MOPS buffer (20 mM, pH 7.0) for quantitation of the relative content of free peptide before (whole) and after (supernatant) centrifugation to sediment the nanosheets.

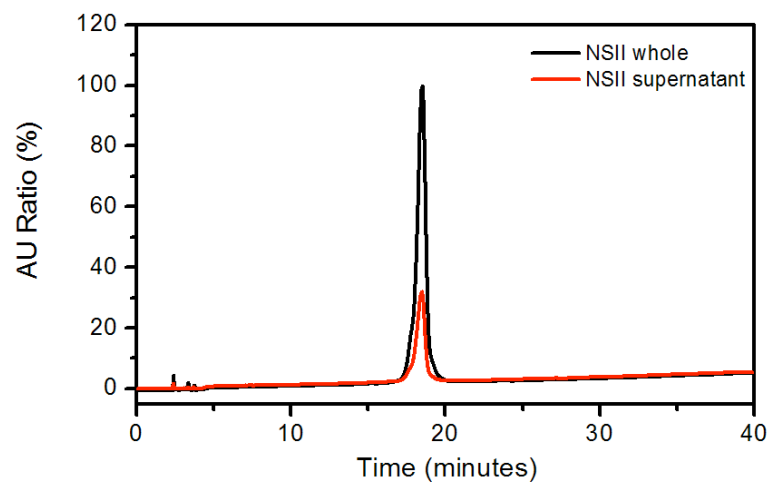


Figure S9. Analytical HPLC analysis of **NSII** (2.5 mg/mL) in MOPS buffer (20 mM, pH 7.0) for quantitation of the relative content of free peptide before (whole) and after (supernatant) centrifugation to sediment the nanosheets.

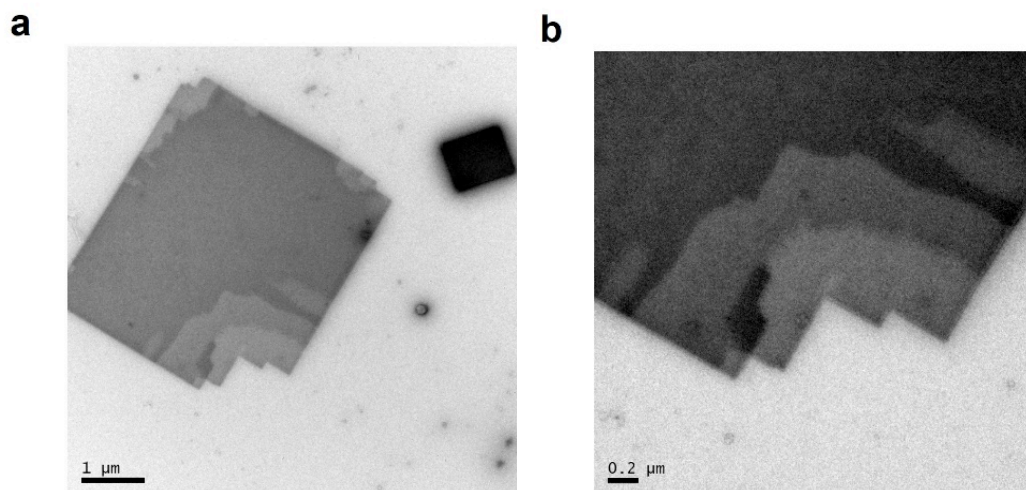


Figure S10. TEM images of **NSI** nanosheets (2 mg/mL) in stages of incomplete growth in MOPS buffer (20 mM, pH 7.0).

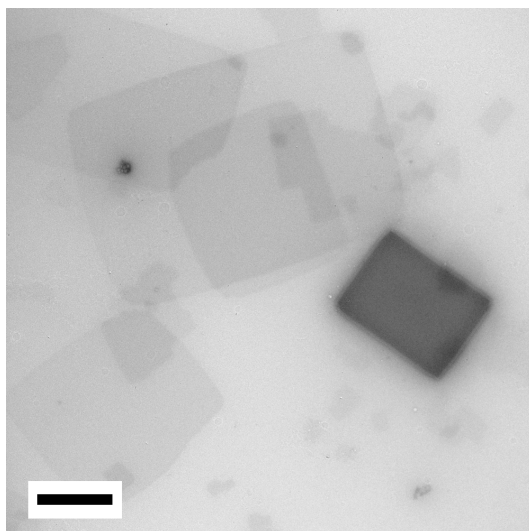


Figure S11. TEM image of nanosheets resulting from self-assembly of an equimolar mixture of NSI (2.0 mg/mL) and NSII (2.5 mg/mL) in MOPS buffer (20 mM, pH 7.0). The nanosheets observed in the mixture were consistent with assemblies observed independently for the individual peptides. Scale bar = 1 μm .

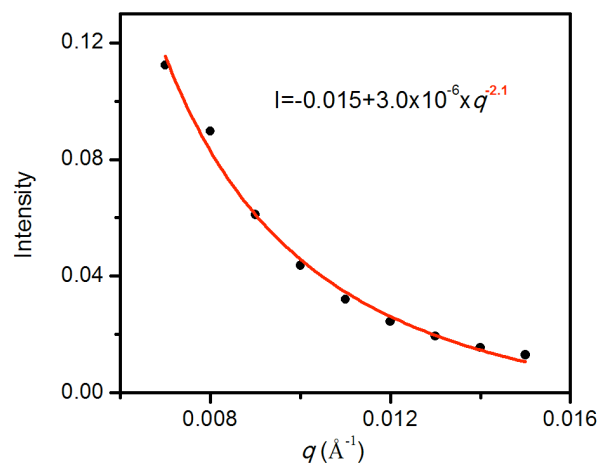


Figure S12. Fit of the SAXS scattering data within the Porod region for nanosheets of **NSI** (4 mg/mL) in MOPS buffer (20 mM, pH 7.0). The mass fractal dimension on the momentum transfer (q) approximates the value expected for sheet-like forms.

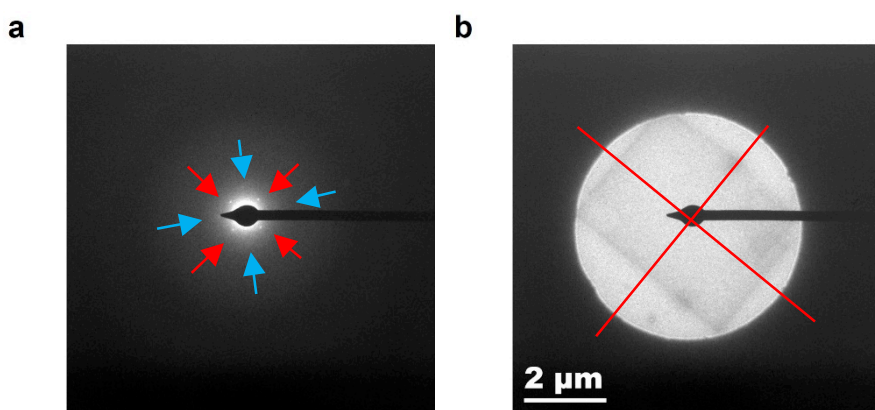


Figure S13. Electron diffraction of individual **NSI** nanosheets. **(a)** Electron diffraction pattern of a representative **NSI** nanosheet comprising major diffraction lattice (red arrows), and weak lattice (blue arrows). **(b)** Corresponding TEM image of the nanosheet under investigation, with the edge directions (red cross) indicating the positions of the major lattice orientations.

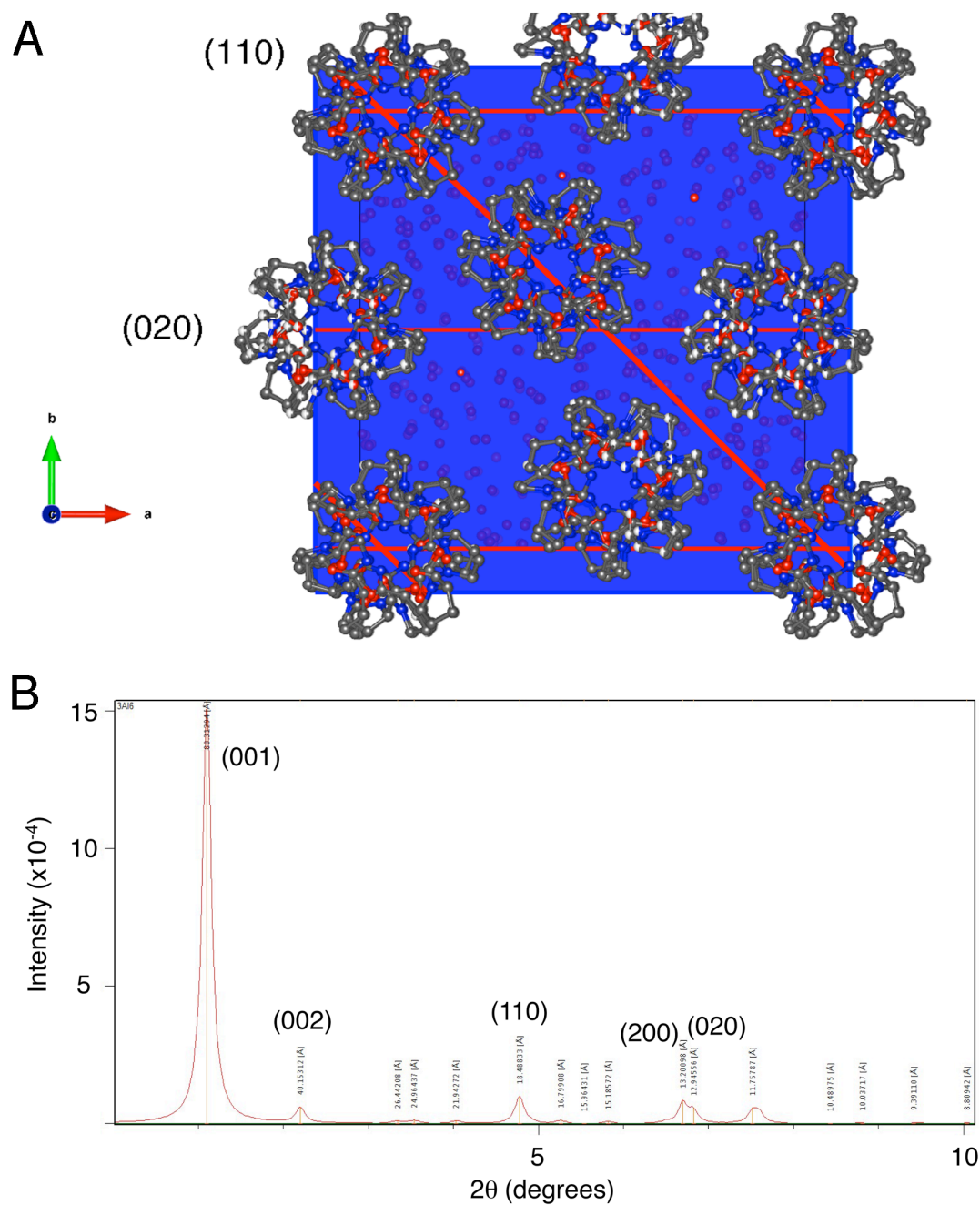


Figure S14. A. View along the *c*-axis of the pseudo-tetragonal packing of triple helices in the *ab*-plane for crystal structure of collagen-mimetic peptide (Pro-Pro-Gly)₉ (PDB ID: 3AI6), indicating the position of the (020) and (110) lattice planes. **B.** Calculated powder pattern from the structure of 3AI6. The strongest reflections in the *ab*-plane correspond to the (200)/(020) and (110) spacings.

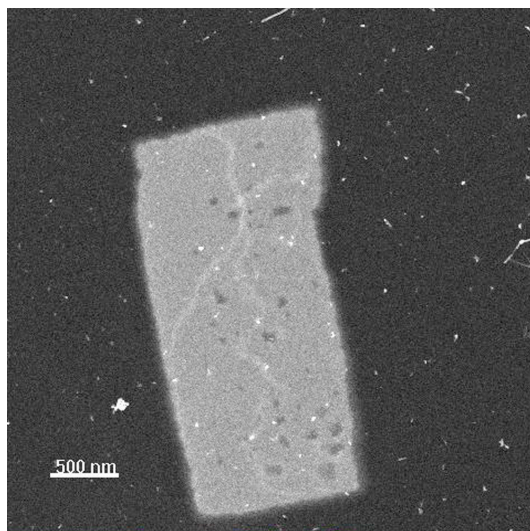


Figure S15. Dark-field STEM of a freeze-dried, unstained specimens of single-layer **NSI** nanosheets (4 mg/mL) assembled in MOPS buffer (20 mM, pH 7.0).

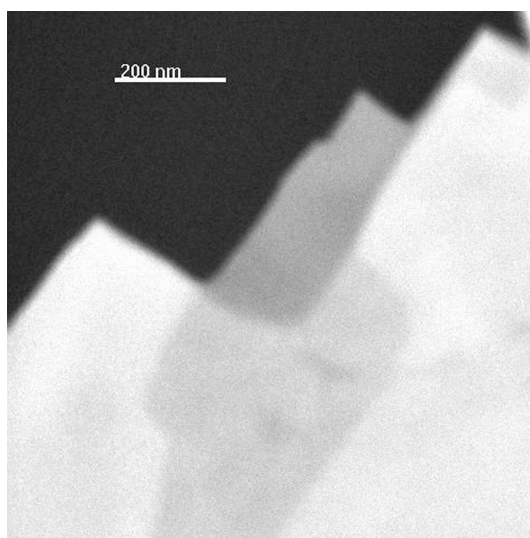


Figure S16. Dark-field STEM of freeze-dried, unstained specimens of a multi-layer nanosheet of **NSI** (4 mg/mL) assembled in MOPS buffer (20 mM, pH 7.0). Multiple terraces are observed on the nanosheet that correspond to differences in the number of packed layers across the surface.

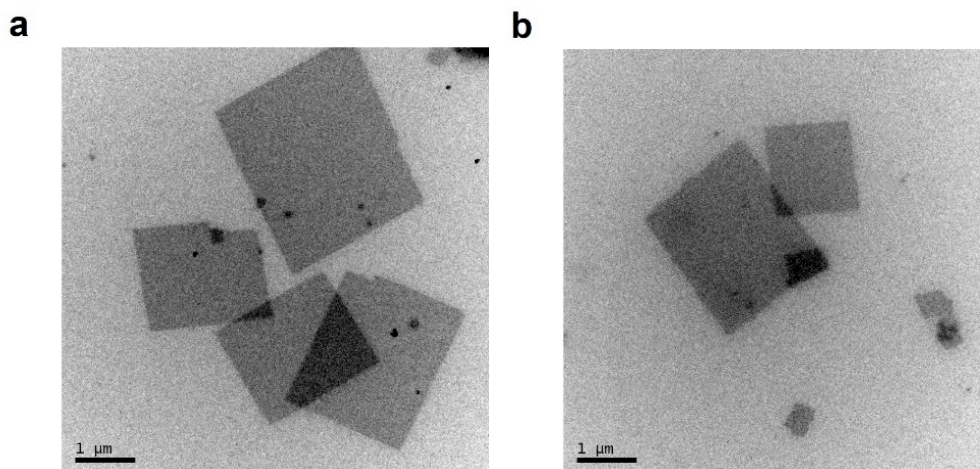


Figure S17. Representative TEM images of **NSII** nanosheets (2.5 mg/mL) that self-assembled from aqueous solutions of MOPS buffer (20 mM, pH 7.0) incubated at 30 °C (a) and at 42 °C (b) for 3 days.

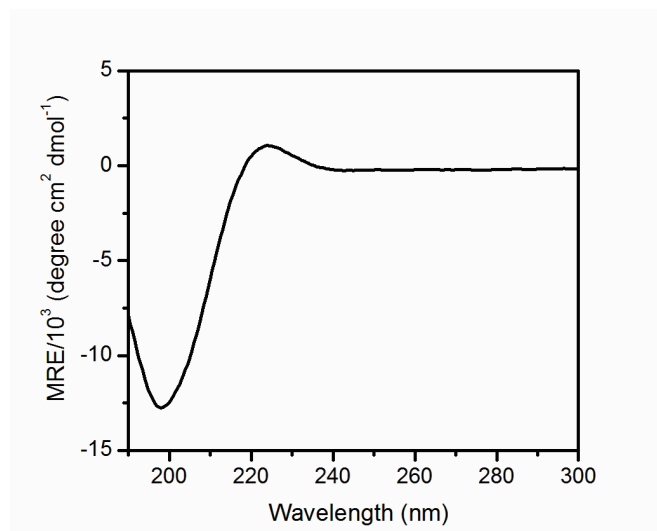


Figure S18. CD spectrum of an aqueous solution of peptide **NSII*** (2.8 mg/mL) in MOPS buffer (20 mM, pH 7.0).

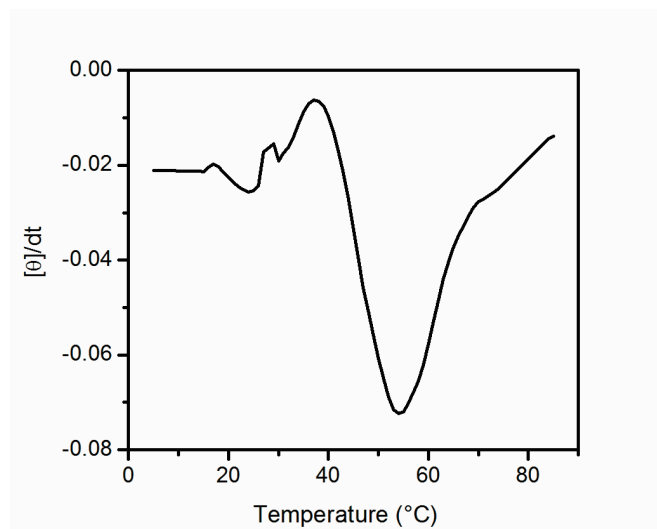


Figure S19. First derivative of the CD melting curve derived from monitoring the signal at 224 nm as a function of temperature for an aqueous solution of **NSII*** in MOPS buffer (20 mM, pH 7.0). A T_m value of 54 °C is estimated from the minimum in the first derivative curve.

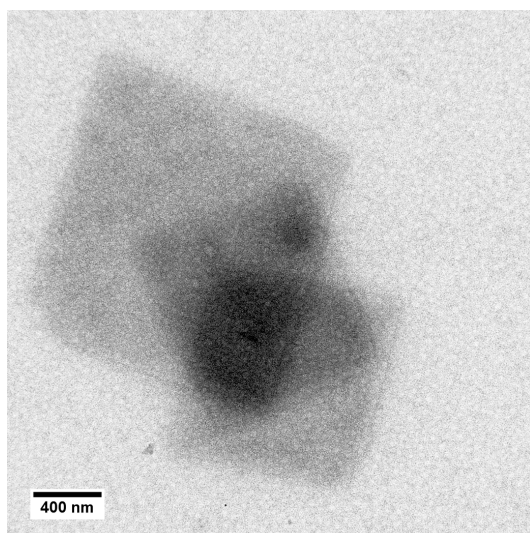


Figure S20. Representative TEM image of **NSII*** nanosheets assembled after a 3 week incubation period from a 10 mg/ml solution in MOPS buffer (20 mM, pH 7.0).

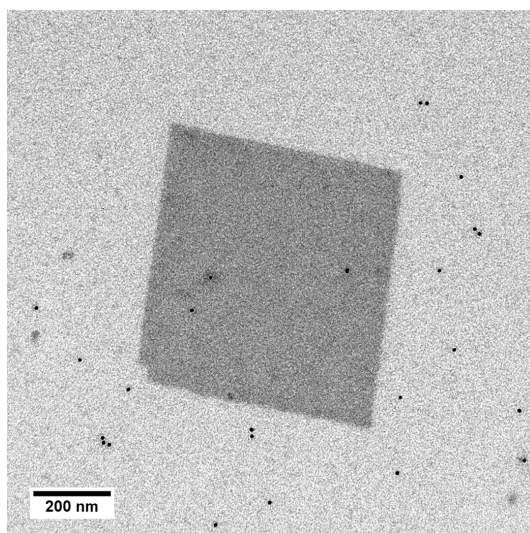


Figure S21. Representative TEM image of the non-biotinylated **NSII** nanosheets after interaction with streptavidin-tagged gold nanoparticles.

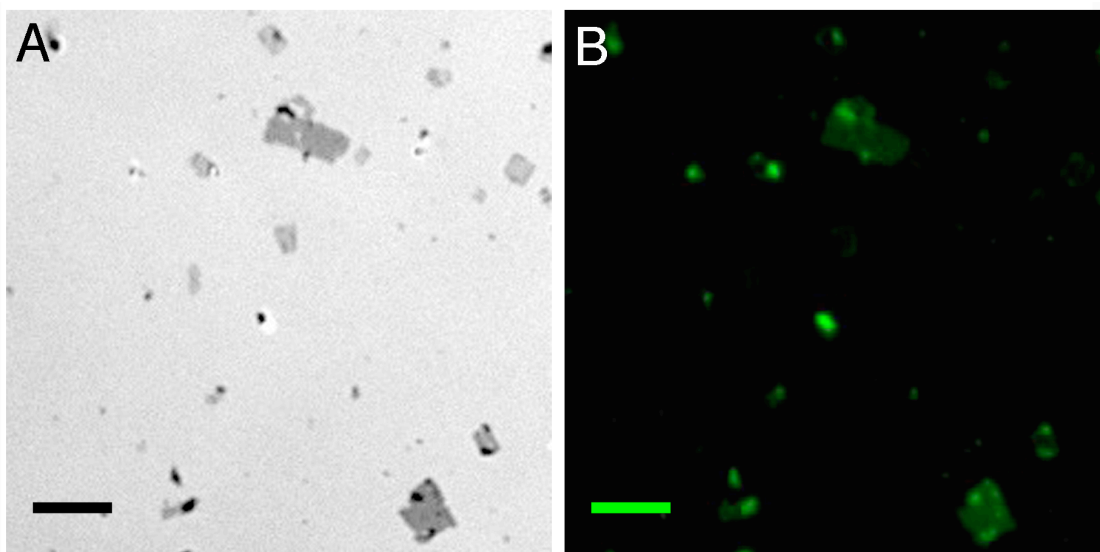


Figure S22. (A) Reflection interference contrast microscopic (RICM) and (B) fluorescence microscopic images of **NSII*** sheets immobilized on a biotinylated glass surface and stained with streptavidin-Alexa488 (scale bar = 4 μm).

Table S1. Calculated intensities and *d*-spacings for selected lattice planes derived from the crystal structure of a collagen-mimetic peptide (PDB ID: 3AI6) with pseudo-tetragonal packing of triple helices.

H	K	L	2θ	Intensity	<i>d</i> (Å)
0	0	1	1.10	10000000	80.162
0	0	2	2.20	364330	40.083
0	0	3	3.31	25590	26.724
1	0	0	3.35	37959	26.357
0	1	0	3.41	0	25.924
1	0	1	3.53	30785	25.043
-1	0	1	3.53	16289	25.043
0	1	1	3.58	15100	24.666
0	-1	1	3.58	15100	24.666
-1	0	2	4.01	3	22.034
1	0	2	4.01	45988	22.023
0	-1	2	4.06	10714	21.763
0	1	2	4.06	10709	21.763
-1	0	3	4.71	1626	18.769
1	0	3	4.71	14605	18.761
0	-1	3	4.75	2361	18.603
0	1	3	4.75	2362	18.603
1	1	0	4.78	319787	18.486
-1	1	0	4.78	319787	18.486
2	0	0	6.71	485232	13.180
0	2	0	6.82	290543	12.960