Supporting information for "Imaging Nanoscale Heterogeneity in Ultrathin Biomimetic and Biological Crystals"

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[a]Region	1643 cm^{-1}		1666 cm^{-1}		1684 cm^{-1}	
	p_0	$\Delta\nu$ (cm^{-1})	p_0	$\Delta\nu$ (cm^{-1})	p_0	$\Delta\nu$ (cm^{-1})
1 (blue)	0.12	22	0.11	30	0.08	16
2 (red)	0.13	28	0.1	31	0.07	16
3 (orange)	0.15	22	0.14	32	0.1	18

[a]See corresponding regions indicated by red, blue, and orange markers in Figure 3a and spectra in Figure 3b.

Table S1. Fit parameters of the triple Gaussian fit to the catalase spectra shown in Figure 3b.

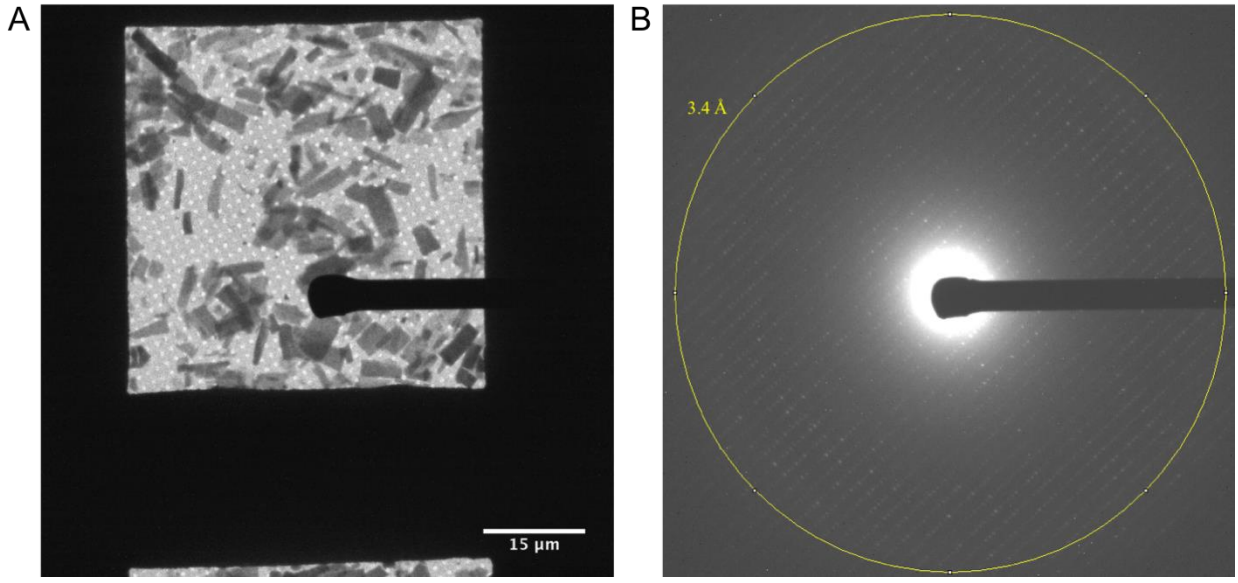


Figure S1. The quality of catalase crystals analyzed by cryo-TEM and electron diffraction. a) Low magnification image of crystals shows a defined distribution of crystals in the micron size range. b) Diffraction pattern of a representative well-ordered catalase crystal with reflections extending to 3.4 Å.

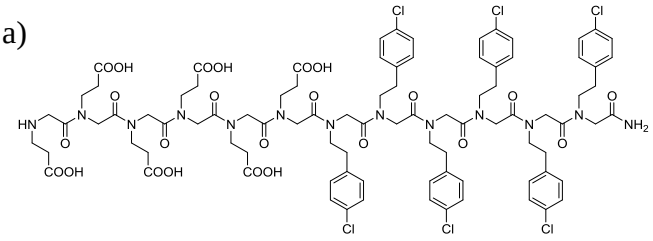
Peptoid synthesis

Methods for the automated solid-phase synthesis

Lipid-like peptoids were synthesized on a commercial Aapptec Apex 396 robotic synthesizer using a modified solid-phase submonomer synthesis method. Rink amide resin (0.09 mmol) was

used to generate C-terminal amide peptoids. In this method, the Fmoc group on the resin was deprotected by adding 2 mL of 20% (v/v) 4-Methylpiperidine/N, Ndimethylformamide (DMF), agitating for 20 min, draining, and washing with DMF. All DMF washes consisted of the addition of 1.5 mL of DMF, followed by agitation for 1 min (repeated five times). An acylation reaction was then performed on the amino resin by the addition of 1.6 mL of 0.6 M bromoacetic acid in DMF, followed by 0.35 mL of 50% (v/v) N, N-diisopropylcarbodiimide (DIC)/DMF. The mixture was agitated for 30 minutes at room temperature, drained, and washed with DMF for 5 times. Nucleophilic displacement of the bromide with various primary amines occurred by a 1.6 mL addition of the primary amine monomer as a 0.6 M solution in N-methyl-2-pyrrolidone (NMP), followed by agitation for 60 minutes at room temperature. The monomer solution was drained from the resin, and the resin was washed with DMF for 5 times. The acylation and displacement steps were repeated until a lipid-like peptoid of the desired length was synthesized.

a)



Pep-1

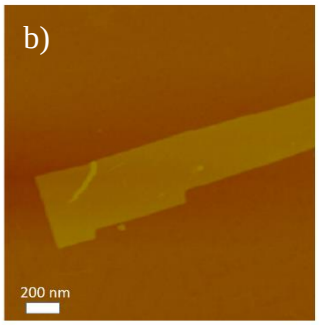


Figure S2. a) Structural of peptoid composing the nanosheets investigated in this study. b) Representative AFM image of a ~4 nm peptoid layer.

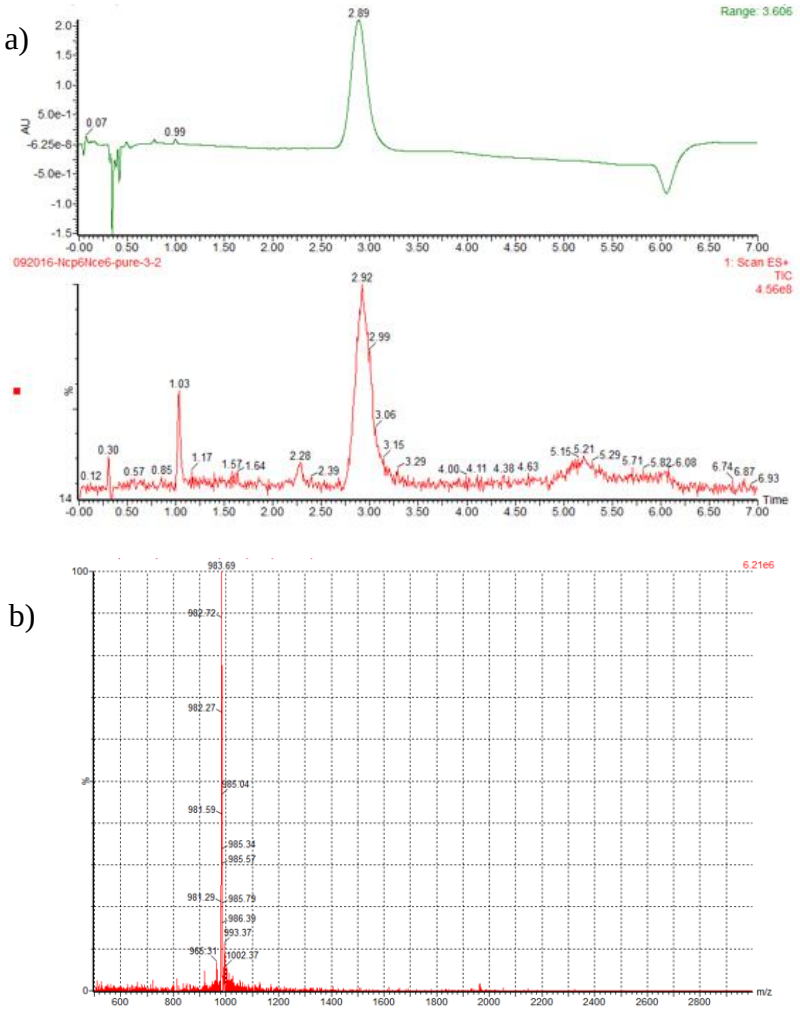


Figure S3. a) Ultra performance liquid chromatography characterization of Pep-1 with the gradient of 5% - 95% CH_3CN in H_2O. b) MS characterization of Pep-1.

Assembly of Pep-1:

1 μmol of lyophilized Pep-1 powder was dissolved in 200 μL of water and acetonitrile (v/v=1:1) mixture to make 5.0 mM clear solution. The mixture was then put in the 4 ^{o}C refrigerator for slow evaporation. Over four days' generation, gel-like materials including a large number of 2D nanosheets was obtained.