Supporting Information

Assembly of Magnetic Nanosheets at the Air-Water Interface by Peptides Inspired by Magnetotactic Bacteria

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Biomineralization*:* For the synthesis of nanosheets 0.12 mg of LHα14 (Ac-LHHLLHLLHHLLHL, M=1788,2 g/mol, Genscript, >90%) was dissolved in 100 µl H_2O and slowly dropped onto the surface of 18.5 ml of H_2O in a temperature-controlled Langmuir trough. After 45 min 1.5 ml of a precursor solution of 0.5 M $FeCl_3 \cdot 6H_2O$ and 0.5 ml of a 0.5 M $FeCl_2 \cdot 4H_2O$ (Sigma-Aldrich, 97%) were injected into the subphase of the trough. The solution was then heated to 65°C for 30 min. The iron oxide precipitation was started by increasing the pH by placing 7 ml of concentrated NH_4OH (VWR Chemicals) next to the trough and covering using a desiccator lid. After 45 min the solution was allowed to cool to RT and then the sample was transferred.

Sum Frequency Generation Spectroscopy: SFG experiments and the laser setup have been described elsewhere.[1] SFG was generated in reflection at an angle of 60° (IR) and 55° (VIS) respective to the surface normal. All data was acquired at 22° C in D_2O as solvent. After background correction, the energy was calibrated using the sum frequency signal originating from the surface of a z-cut quartz crystal. The adsorption of peptides was followed by surface tension measurements using the Wilhelmy-method using a Kibron Inc., Delta Pi instrument. As can be seen in Figure S1, the surface adsorption and assembly stabilizes after approximately 3000 s. The initial rise of the surface tension above the equilibrium value is likely explained by reorganization and assembly of the peptide monolayer.

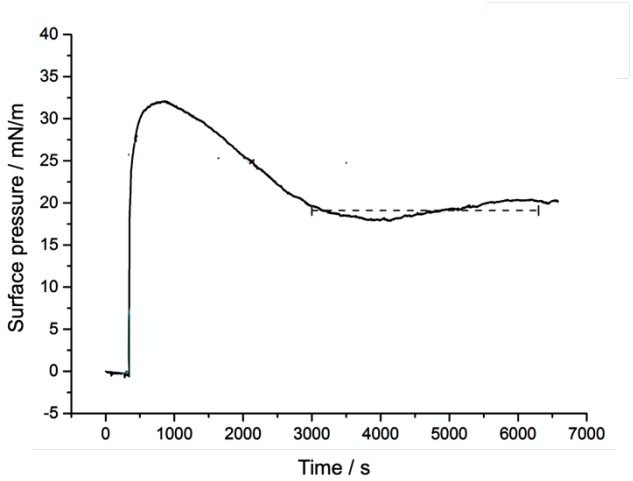

Figure S1: Surface pressure recorded as a function of time after injection of the LHα14 peptide. The reported surface pressure is normalized to the value of pure water. The adsorption equilibrates after approximately 3000 s. The SFG experiments and mineralization steps are conducted in the timeframe between 3000 s and 6000 s.

X-ray photoelectron spectroscopy: XPS data were collected on a Kratos AXIS Ultra DLD spectrometer. The instrument uses a monochromatic Al Kα X-ray source. The electron take-off angle (between the sample surface plane and the axis of the analyzer lens) was 90° using small spot analysis detection mode with a spot size of 100 μm^2. The scans were acquired with an analyzer pass energy of 80 eV. The base pressure in the analysis chamber was better than $5 \cdot 10^{-9}$ mbar. The data analysis was performed using the Vision Processing software.

Electron Microscopy*:* Nano-sheets were transferred from the air-water interface onto silicon wafers via the Schaefer technique. In order to allow reproducible transfers of the 2D sheets, we use a homebuilt setup equipped with micrometer screws. The setup is depicted in Figure S2.

These samples were mounted on aluminum stubs and analyzed with a Zeiss Gemini 1530 SEM instrument and an accelerating voltage of 750 V. TEM samples were lifted off the solution surface using TEM grids. The experiments were carried out with JEM 1400, JEOL Ltd., Japan, and operating voltage of 120 kV was applied during measurement. Micrographs were recorded on a Gatan Ultrascan CCD Camera.

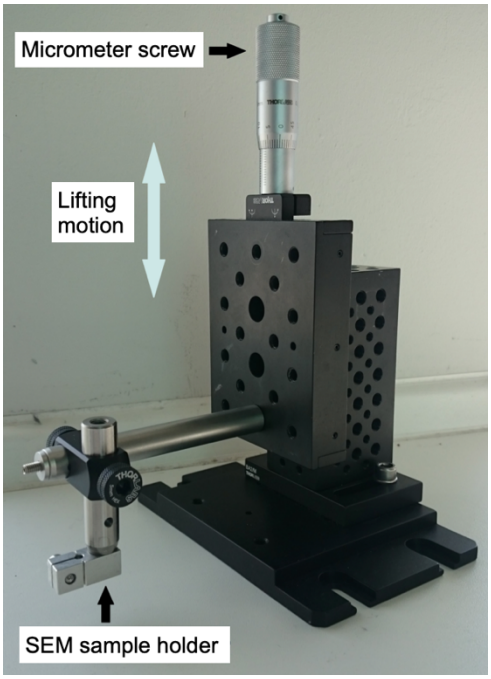

Figure S2: The Langmuir-Schäfer transfer requires that the sample holder can be brought into contact with the sample surface parallel to the TEM grid surface. This setup allows reproducible approach and retraction from the solution surface. Using the micrometer screw, the sample was brought in contact with the solution surfaces. Then the sample was rinsed with MilliQ-water and the sample was removed from the holder, dried in a stream of nitrogen and in nitrogen-backfilled containers until analysis.

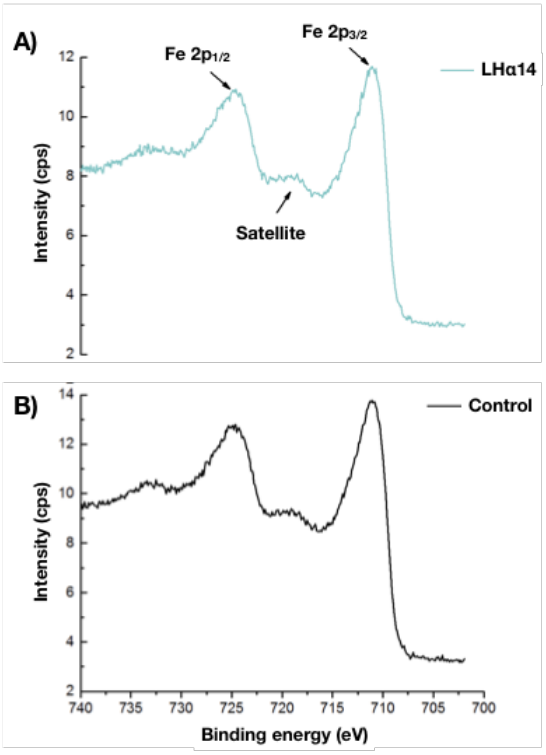

Figure S3: High resolution Fe XPS spectra of the iron oxide films. A) Fe 2p XPS spectra for the iron oxide nanosheets generated by LHα14. B) Fe 2P XPS spectra for the negative control film precipitated without peptides.

1. T. W. Golbek, L. Schmüser, M. H. Rasmussen, T. B. Poulsen and T. Weidner, *Langmuir*, 2020, **36**, 3184-3192.