Supporting Information

Hauser et al. 10.1073/pnas.1014796108

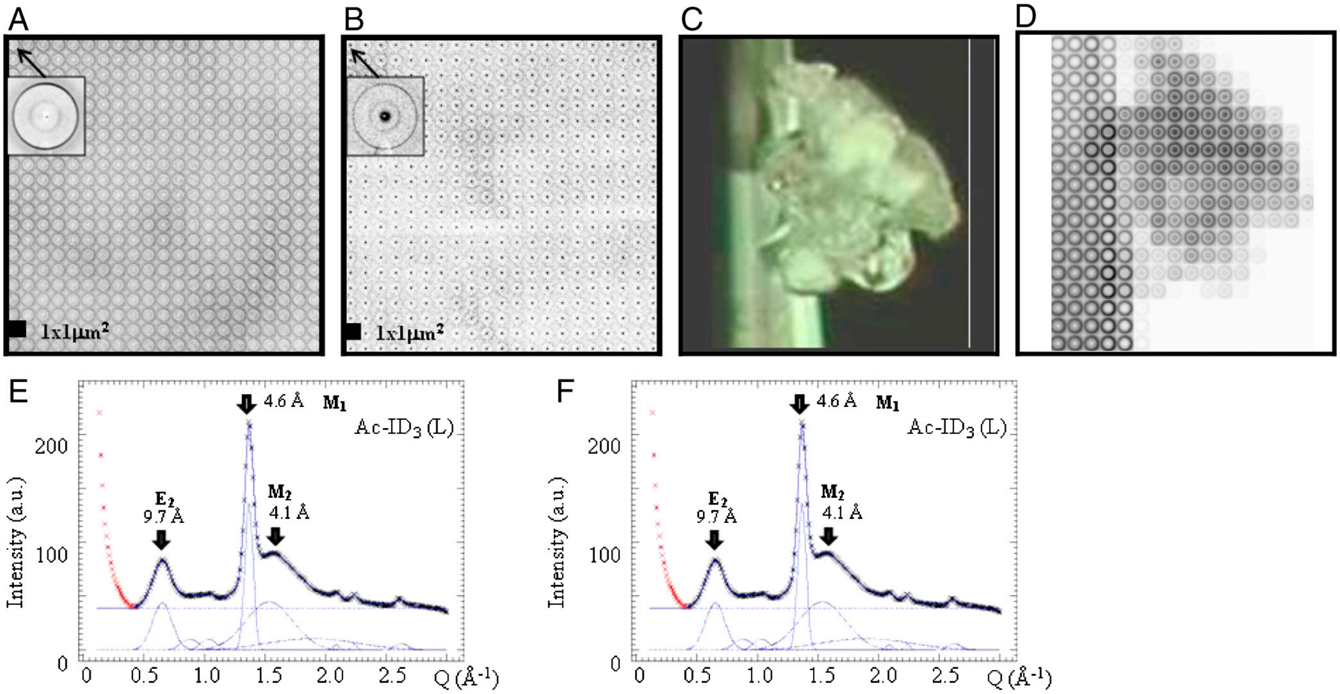
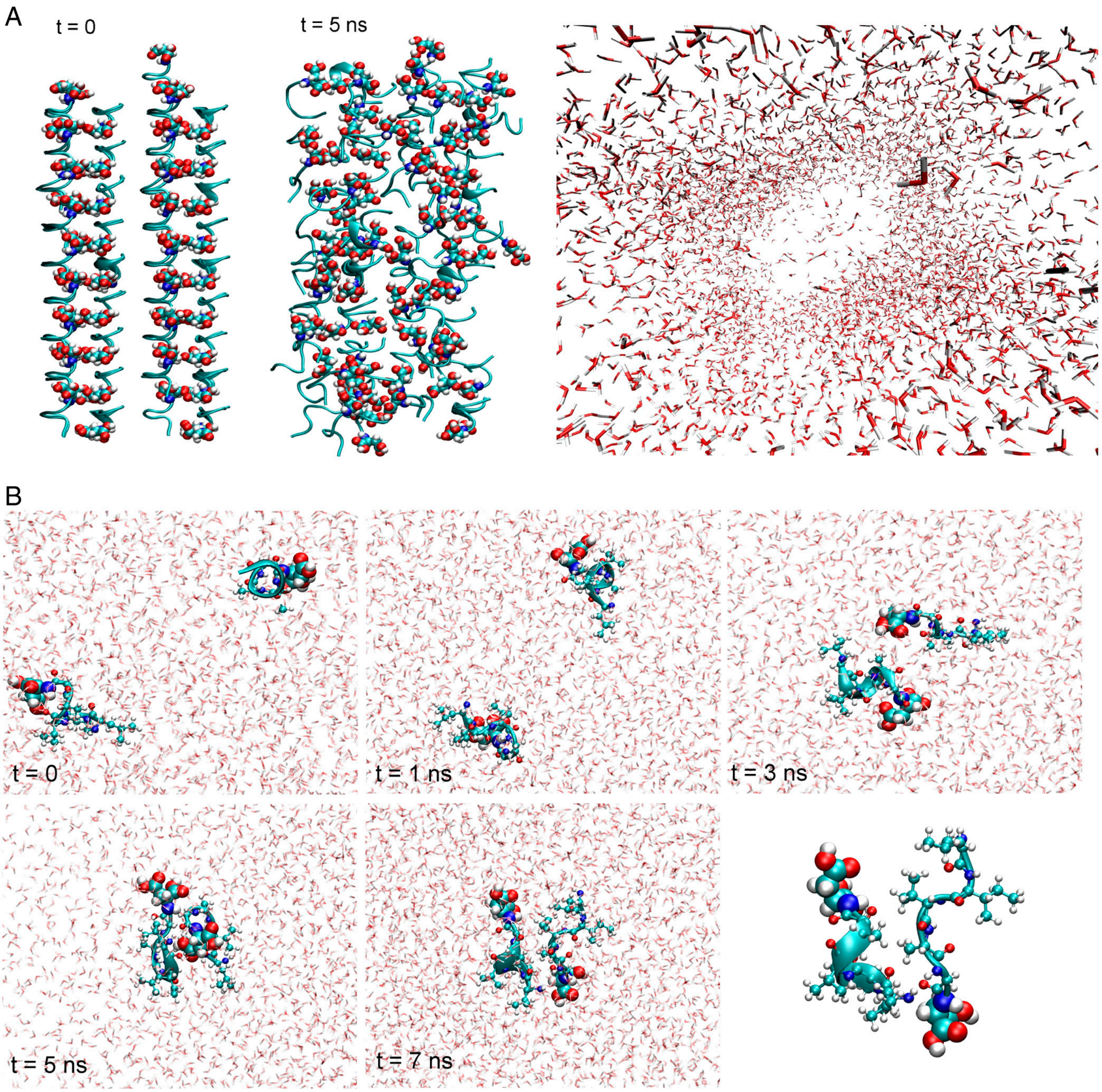


Fig. S1. (*A* and *B*) Composite images from 1 ∗ 1 μm^2 step-resolution raster-microdiffraction for Ac-ID$_3$ (*A*) and Ac-LD$_6$ (*B*) hydrogel samples at 100 K. Individual patterns are shown as inset. Intensity fluctuations in the patterns are due to variable sample thickness. (*C*) Optical microscopy images of Ac-LD$_6$ residuals attached to glass capillaries. (*D*) Composite images from raster microdiffraction with 20 ∗ 20 μm^2 (Ac-LD$_6$) step-resolution. Fiber texture is observed. (*E* and *F*) Azimuthally averaged patterns of Ac-ID$_3$ (*E*) and Ac-LD$_6$ (*F*) at 100 K. The patterns have been fitted by Gaussian profiles for the Bragg peaks and a 0-order polynomial for the residual background (in blue). Positions of the individual peaks are indicated by arrows. The part of the pattern excluded to the fit is in red. Small amounts of residual ice are indicated.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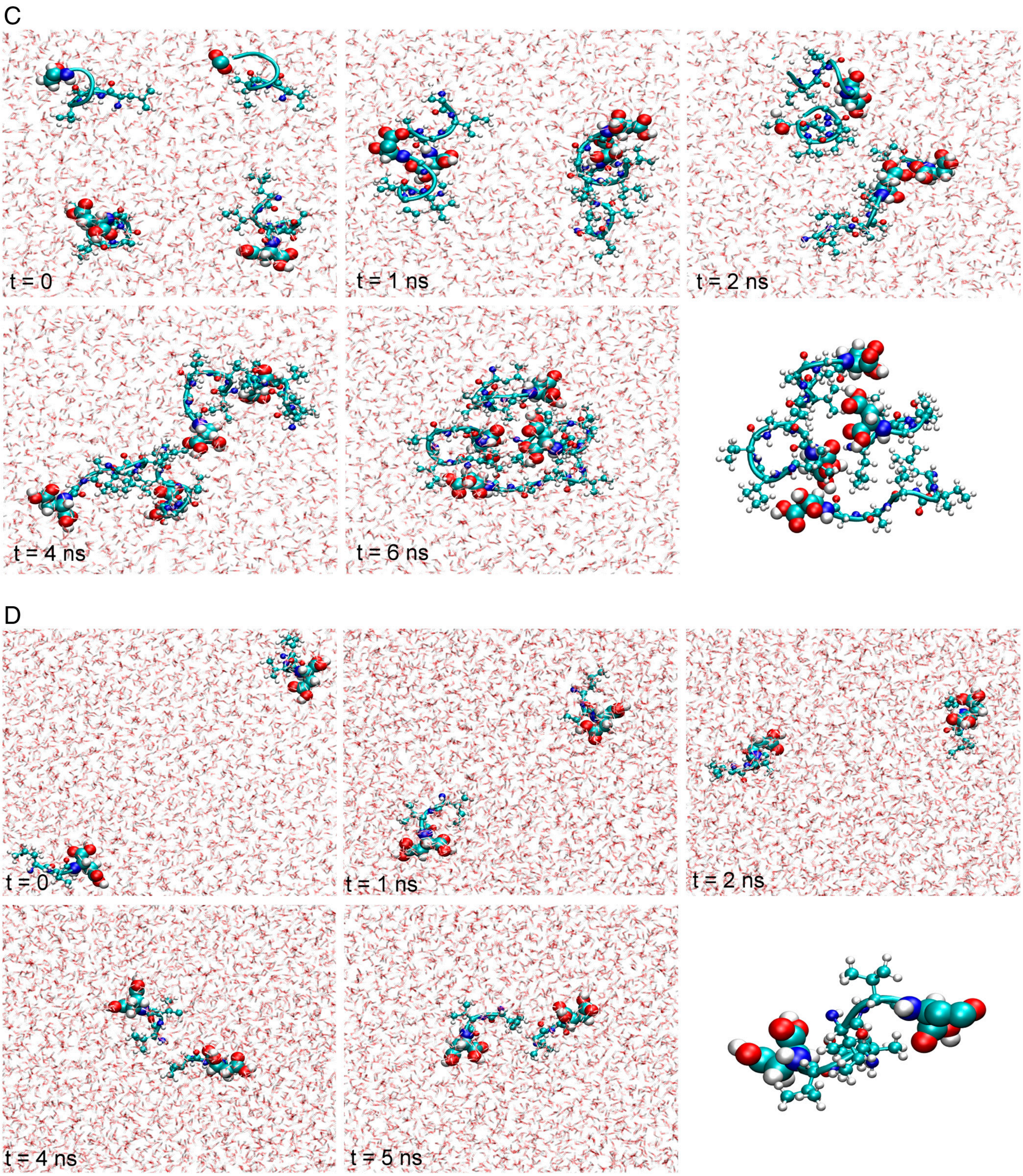
t = 5 ns
B
t = 0
t = 1 ns
t = 3 ns
t = 5 ns
t = 7 ns


C
t = 0
t = 1 ns
t = 2 ns
t = 4 ns
t = 6 ns
D
t = 0
t = 1 ns
t = 2 ns
t = 4 ns
t = 5 ns

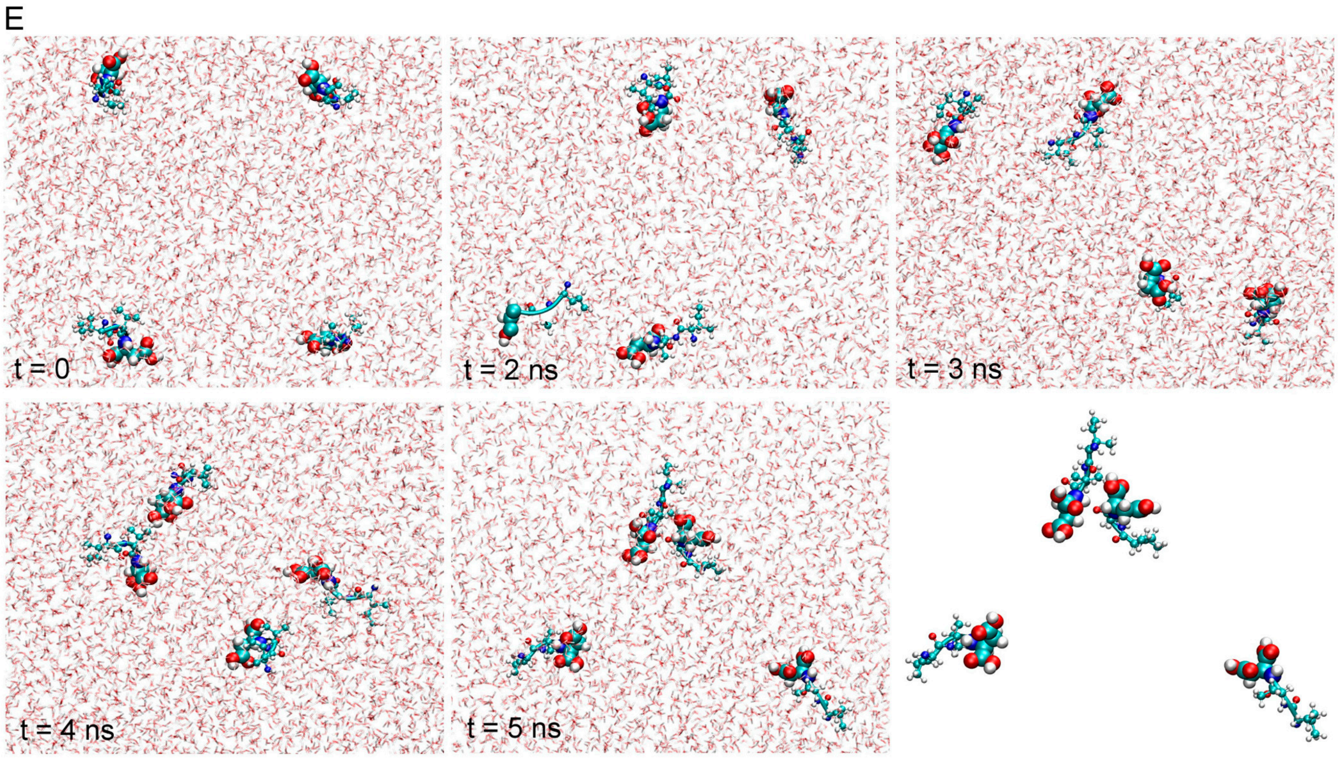


Fig. S2. (*A*) Snapshots from molecular dynamics simulations of 72 hexamer Ac-LD$_6$ peptides stacked vertically in 5025 water molecules. Peptides are initially paired in an antiparallel orientation. The pairs are stacked vertically twice in both the *x* and *y* direction, and nine times in the *z* direction, resulting in a fiber consisting of 72 peptides (top left). This configuration can be compared to the one shown in the main paper (Fig. 3*B*), where the peptides are stacked horizontally. After 5 ns of simulations, the fiber remains intact. Peptides are much more disordered in this configuration than in the fiber with the peptides stacked horizontally (top right). The water penetration into the peptide is minimal, with most of the water molecules forming a layer around the fiber (right). (*B*) Snapshots from the simulation of two hexamer Ac-LD$_6$ peptides in 2,107 molecules of water shown at $t = 0$, 1 ns, 3 ns, 5 ns, and 7 ns. Initially the peptides are placed apart from each other. The peptides gradually move closer to each other and by $t = 3$ ns, the peptides have formed a pair. The peptides remain in the pair formation throughout the rest of the simulation until $t = 7$ ns, indicating the stability of the pair configuration. In the final structure, the pairs are in an antiparallel orientation to each other. However, more detailed analysis is required to confirm if the antiparallel orientation is the preferred orientation. To this end, we have conducted a DFT calculation (1) [M06-2X (2) /6-31G (3)] of two Ac-LD$_6$ peptides in the parallel and antiparallel orientation, respectively. Our preliminary results indicate a strong preference for the antiparallel orientation by 82.4 KJ/mol. (*C*) Snapshots from the simulation of four hexamer Ac-LD$_6$ peptides in 2,036 molecules of water shown at $t = 0$, 1 ns, 2 ns, 4 ns, and 6 ns. Initially the peptides are placed apart from each other. Here the peptides form pairs very quickly, and by $t = 1$ ns, two pairs of peptides have formed. The pairs are stable and gradually come closer to each other. By $t = 6$ ns, a cluster of four peptides has formed. (*D*) Snapshots from the simulation of two trimer Ac-ID$_3$ peptides in 6226 molecules of water shown at $t = 0$, 1 ns, 2 ns, 4 ns, and 5 ns. To ascertain whether the tendency to form pairs is a general behavior of the peptides in water, we also performed simulations of the shorter Ac-ID$_3$ peptides. Initially two peptides are placed apart from each other. The peptides gradually come closer, and begin to form a pair by $t = 4$ ns. The peptides remain in the pair formation for the rest of the simulation. The dynamics of the pair formation is slower compared to the longer Ac-LD$_6$ peptide, most likely due to the relatively lower hydrophobicity of the shorter Ac-ID$_3$ peptide. However, the tendency to form pairs in water is consistent with the Ac-LD$_6$ peptide. (*E*) Snapshots from the simulation of four trimer Ac-ID$_3$ peptides in 6,181 molecules of water shown at $t = 0$, 2 ns, 3 ns, 4 ns, and 5 ns. As for the case of the two Ac-ID$_3$ peptides, the dynamics of the pair formation is slower than for the hexamer Ac-LD$_6$ peptide. By the end of the simulation at $t = 5$ ns, only two of the peptides have formed a pair. However the peptides have moved closer to each other compared to their initial positions. The peptides will most likely form a cluster of four peptides if the simulation time were to be extended. This is an attribute to the lower hydrophobicity of the shorter Ac-ID$_3$ compared to the Ac-LD$_6$.

1 Frisch MJ et al. (2009) Gaussian 09, Revision A.02. (Gaussian Inc., Wallingford, CT).
2 Zhao Y, Truhlar DG (2008) The M06 suite of density functionals for main group thermochemistry, thermochemical kinetics, noncovalent interactions, excited states, and transition elements: Two new functionals and systematic testing of four M06-class functionals and 12 other function. *Theoretical Chemistry Accounts* 120:215–241.
3 Hehre WJ, Radom P, Schleyer PvR, Pople JA (1986) *Ab Initio Molecular Orbital Theory* (Wiley, New York).