Electronic Supplementary Information (ESI)

**Force field parameters**

The total bond energy is the sum of four contributions:

\[ U_{\text{bond}} = U_l + U_\phi + U_\theta + U_\lambda, \]

where

\[ U_l = \frac{k_l}{2} (l - l_0)^2 \]

\[ U_\phi = \frac{k_\phi}{2} (\phi - \phi_0)^2 \]

\[ U_\theta = 1 + a_2 \theta^2 + \theta^4 + a_6 \theta^6 \]

\[ U_\lambda = \frac{k_\lambda}{2} (|\lambda| - \lambda_0)^2 \]

where \( l \) and \( \phi \) denote bond length and bond angle, respectively. Dihedral angles (see Fig. 1a in the main text) are denoted by \( \lambda \) and \( \theta \). The parameters of the force field parameters are given in Table 1. The values of \( a_2 \) and \( a_6 \) were computed for each \( \theta_0 \) to get the potential with minima at \( \pm \theta_0 \) and a barrier height of \( \Delta E = 5/2 \).

<table>
<thead>
<tr>
<th>term</th>
<th>beads affected</th>
<th>parameters</th>
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</thead>
<tbody>
<tr>
<td>( U_{lBS} )</td>
<td>( B - S )</td>
<td>( k_l = 50, l_0 = 2 )</td>
</tr>
<tr>
<td>( U_{lBB} )</td>
<td>( B - B )</td>
<td>( k_l = 100, l_0 = 1 )</td>
</tr>
<tr>
<td>( U_{\phiBB} )</td>
<td>( S - B - B )</td>
<td>( k_\phi = 200, \phi_0 = 90^\circ )</td>
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<tr>
<td>( U_{\thetaBB} )</td>
<td>( B - B - B )</td>
<td>( k_\theta = 50, \phi_0 = 180^\circ )</td>
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<tr>
<td>( U_\lambda )</td>
<td>( S - B, B_1 + 2 - S )</td>
<td>( k_\lambda = 10 )</td>
</tr>
<tr>
<td>( U_\theta )</td>
<td>( S1 - B, B_1 + 1 - S2 )</td>
<td>( a_2 ) and ( a_6 ) - see the text</td>
</tr>
<tr>
<td>LJ</td>
<td>( B )</td>
<td>( \varepsilon = 1, \sigma_B = 4 )</td>
</tr>
<tr>
<td>LJ</td>
<td>( S1, S2 )</td>
<td>( \varepsilon = 1, \sigma_S = 1 )</td>
</tr>
</tbody>
</table>

**Simulations**

Simulations were run with the LAMMPS package in three stages:

1. Short equilibration of side strands with backbone beads’ location restrained: timestep=0.5 × 10^{-3}t_D, total time=400t_D

2. Short molecular dynamics NVT run at \( T^* = 1.5 \) in order to relax the initial configuration run to change the initial configuration of the fibrils: timestep=10^{-2}t_D, total time=800t_D

3. Langevin dynamics: timestep=10^{-2}t_D, total time=10^6t_D

**Helix angle**

As illustrated in Fig. 1, the helix angle, \( \alpha_0 \) is the angle between the helical curve and the vertical axis. The arc \( a \) is spanned by horizontal vectors \( \rho_1 \) and \( \rho_2 \) with \( \lambda_0 \) being the angle between them. Next, \( b \) is the axial distance between the two consecutive side strand beads. The length of arc \( a \) is therefore \( \rho_\lambda \), where \( \rho \) corresponds to \( l_{BS} = 2 \), whereas \( b \) is \( b = 2l_{BB} = 2 \). Hence, \( \tan(\alpha_0) = \lambda_0 \) and \( \alpha_0 = \arctan(\lambda_0) \).

**Relation of the chirality, \( \chi \), to the torsion of the chain**

Below we show that in the limit of a continuous curve Eq. (2) gives the local torsion of the chain. Consider a curve...
parametrized by its arc length, \( s \), and three consecutive tangent vectors \( t_\pm = t(-\epsilon), \ t = t(0) \) and \( t_\mp = t(\epsilon) \), separated by a distance \( \epsilon \) along the arc length. The tangent vectors, together with the corresponding normal and binormal vectors constitute a local orthogonal trihedron fulfilling the Frenet-Serret relations

\[
\begin{align*}
\mathbf{t}' &= \kappa \mathbf{n} \\
\mathbf{n}' &= -\kappa \mathbf{t} + \tau \mathbf{b} \\
\mathbf{b}' &= -\kappa \mathbf{t}
\end{align*}
\]

where \( \kappa \) is the curvature and \( \tau \) is the torsion of the curve. The prime denotes differentiation with respect to the arc length.

For small \( \epsilon \)

\[
\mathbf{t}_\pm = \mathbf{t} \pm \mathbf{t}' \epsilon + \frac{1}{2} \mathbf{t}'' \epsilon^2 + \ldots
\]

Using Frenet-Serret formulas

\[
\mathbf{t}_\pm = \mathbf{t} \pm \kappa \epsilon \mathbf{b} + \frac{1}{2} \epsilon^2 (\kappa' \mathbf{n} - \kappa^2 \mathbf{t} + \kappa \tau \mathbf{b}) + O(\epsilon^3)
\]

which leads to

\[
\mathbf{t}_- \times \mathbf{t} = \kappa \epsilon \mathbf{b} + \frac{1}{2} \epsilon^2 (\kappa \tau \mathbf{n} - \kappa' \mathbf{b})
\]

and

\[
\mathbf{t} \times \mathbf{t}_+ = \kappa \epsilon \mathbf{b} - \frac{1}{2} \epsilon^2 (\kappa \tau \mathbf{n} - \kappa' \mathbf{b})
\]

Finally

\[
(\mathbf{t}_- \times \mathbf{t}) \cdot (\mathbf{t} \times \mathbf{t}_+) = \kappa^2 \epsilon^2 + \ldots
\]

and

\[
(\mathbf{t}_- \times \mathbf{t}) \cdot \mathbf{t}_+ = \kappa^2 \tau \epsilon^3 + \ldots
\]

The argument of the arc tangent function in Eq. (2) is the ratio of these two terms. Thus, in the limit of small \( \epsilon \) the expression

\[
\arctan^2 \left( \frac{\| \mathbf{v}_i \| \mathbf{v}_{i-1} \cdot (\mathbf{v}_i \times \mathbf{v}_{i+1})}{(\mathbf{v}_{i-1} \times \mathbf{v}_i) \cdot (\mathbf{v}_i \times \mathbf{v}_{i+1})} \right)
\]

is proportional to the local (signed) torsion of the chain.