SUPPLEMENTARY MATERIAL. Computation of the reduced quadratic energy

A. GEOMETRY OF HELICES IN 3D

A rod with arc length $s \in [0, L]$ is defined by a centreline $\mathbf{r}(s)$ and a unit vector field $\mathbf{d}_1(s)$ perpendicular to the unit tangent $\mathbf{r}'(s) =: \mathbf{d}_3(s)$ (prime denotes the s-derivative). A local orthonormal basis of directors is obtained by defining $\mathbf{d}_2(s) := \mathbf{d}_3(s) \times \mathbf{d}_1(s)$ so that $\mathbf{d}_i' = \mathbf{u} \times \mathbf{d}_i, \;\; i = 1, 2, 3$ where $\mathbf{u}$ is the Darboux vector. Any vector, and in particular $\mathbf{u}$, can be written as $\mathbf{u} = \mathsf{u}_1\mathbf{d}_1 + \mathsf{u}_2\mathbf{d}_2 + \mathsf{u}_3\mathbf{d}_3$ with the three components assembled into a triple $\boldsymbol{\mathsf{u}} = (\mathsf{u}_1, \mathsf{u}_2, \mathsf{u}_3)$ (note the use of sans-serif fonts in the director basis).

By defining a resultant force $\mathbf{n}(s)$ and moment $\mathbf{m}(s)$ acting across the cross section at $\mathbf{r}(s)$, and in the absence of distributed body forces and couples, balance of force and moment yields the equilibrium equations

$$\boldsymbol{\mathsf{n}}' + \boldsymbol{\mathsf{u}} \times \boldsymbol{\mathsf{n}} = \mathbf{0}, \tag{1}$$

$$\boldsymbol{\mathsf{m}}' + \boldsymbol{\mathsf{u}} \times \boldsymbol{\mathsf{m}} + \boldsymbol{\mathsf{v}} \times \boldsymbol{\mathsf{n}} = \mathbf{0}, \;\; \boldsymbol{\mathsf{v}} := (0, 0, 1)^T. \tag{2}$$

The general material response of a rod with quadratic energy is given by a linear constitutive relation with eight constants

$$\boldsymbol{\mathsf{m}} = \mathbf{K}(\boldsymbol{\mathsf{u}} - \hat{\boldsymbol{\mathsf{u}}}), \quad \mathbf{K} = \begin{pmatrix} K_1 & 0 & K_{13} \\ 0 & K_2 & K_{23} \\ K_{13} & K_{23} & K_3 \end{pmatrix}, \; K_1 \leq K_2, \tag{3}$$

where $\hat{\boldsymbol{\mathsf{u}}}$ are the Darboux components of the unstressed shape, and $\mathbf{K}$ is positive definite.

Uniform configurations of a filament are solutions with $\boldsymbol{\mathsf{u}}$ constant, so that $\mathbf{u}$ is itself also constant, which implies that uniform configurations have helical centrelines with axis along $\mathbf{u}$ [1] and with curvature, torsion and radius given by $\kappa = \sqrt{\mathsf{u}_1^2 + \mathsf{u}_2^2}$, $\tau = \mathsf{u}_3$ and $R = \kappa/|\boldsymbol{\mathsf{u}}|^2$. Let $\mathbf{e} = \epsilon\mathbf{u}/|\mathbf{u}|$ be a unit vector along the axis such that $\mathbf{e} \cdot \mathbf{r}'(s) \geq 0$, and $\epsilon = +1$ for right-handed helices (and circular arcs) with $\mathsf{u}_3 = \tau \geq 0$ ($\epsilon = -1$, for left-handed helices). The pitch per unit arc length along $\mathbf{e}$ is $p = |\mathsf{u}_3|/|\boldsymbol{\mathsf{u}}|$ while the *coiling angle per unit arc length* is $\rho = |\boldsymbol{\mathsf{u}}|$, so that the number of helical periods is $L\rho/2\pi$ for a helix segment of arc length L. When one helical configuration is deformed into another, it will overwind if $\theta := \rho_2 - \rho_1 > 0$ and unwind if $\theta < 0$ (respectively, increasing or decreasing the number of helical repeats for a given arc length).

Uniform equilibria are characterized by constant triples $\boldsymbol{\mathsf{n}}$, $\boldsymbol{\mathsf{m}}$ and $\boldsymbol{\mathsf{u}}$ that satisfy the equilibrium equations (1-2) and constitutive relations (3). The third component of (2) reads

$$\mathcal{Q}(\boldsymbol{\mathsf{u}}) := \mathsf{u}_1\mathsf{m}_2 - \mathsf{u}_2\mathsf{m}_1 = 0. \tag{4}$$

Together with the constitutive relations (3), $\mathcal{Q}(\boldsymbol{\mathsf{u}})$ defines a quadric in Darboux space that contains the entire u_3-axis and which, apart from degenerate cases (see below), is a one-sheeted hyperboloid that contains all values of $\boldsymbol{\mathsf{u}}$ corresponding to helical equilibria. The origin $\boldsymbol{\mathsf{u}} = 0$ is one 'helical' equilibrium with a straight centreline and force $\boldsymbol{\mathsf{n}}$ of arbitrary magnitude aligned with the centreline. All points on the hyperboloid off the u_3-axis represent helical equilibria with $\kappa > 0$ and unique associated stresses $\boldsymbol{\mathsf{n}}$ and $\boldsymbol{\mathsf{m}}$. Indeed, for $\boldsymbol{\mathsf{u}} \neq 0$, (2) and $\mathcal{Q}(\boldsymbol{\mathsf{u}}) = 0$ imply that $\boldsymbol{\mathsf{n}} = \mu\boldsymbol{\mathsf{u}}$ with $\mu = \mathsf{m}_3 - \mathsf{u}_3(\mathsf{u}_2\mathsf{m}_1 + \mathsf{u}_1\mathsf{m}_2)/(2\mathsf{u}_1\mathsf{u}_2)$ (valid for $\mathsf{u}_1 = 0$ or $\mathsf{u}_2 = 0$, but not both). As the constitutive relations (3) provide $\boldsymbol{\mathsf{m}}$ as a function of $\boldsymbol{\mathsf{u}}$, we now have the basic quantities associated with a helical equilibrium explicitly written as a function of points $\boldsymbol{\mathsf{u}}$ in Darboux space lying on the hyperboloid $\mathcal{Q}(\boldsymbol{\mathsf{u}}) = 0$ and off the u_3-axis.

Conjugate to the two generalized displacements (p, ρ), we need two generalized loads given by the axial components of force and moment. These are the four accessible effective quantities in a single molecule tweezer experiment, and are also entirely natural variables in macroscopic systems. The axial component of the force is $N := \boldsymbol{\mathsf{n}} \cdot \boldsymbol{\mathsf{e}} = \epsilon\mu|\boldsymbol{\mathsf{u}}| =: \mathcal{N}(\boldsymbol{\mathsf{u}})$, while the axial component of the moment is $M := \boldsymbol{\mathsf{m}} \cdot \boldsymbol{\mathsf{e}} = \epsilon\boldsymbol{\mathsf{u}} \cdot \boldsymbol{\mathsf{m}}/|\boldsymbol{\mathsf{u}}| =: \mathcal{M}(\boldsymbol{\mathsf{u}})$. Together, (N, M) forms a wrench: a pure axial force $N\mathbf{e} = \mathbf{n}(L)$ and pure axial moment $M\mathbf{e} = \mathbf{m}(L) - N\mathbf{r}(L) \times \mathbf{e}$ applied at the axis of the helical equilibrium (cf. Fig. 3 in Main Text).

We now have a purely geometric problem; for a given wrench (N, M) the observed response (p, ρ) is determined by the intersection of the level sets of the functions $\mathcal{N}(\boldsymbol{\mathsf{u}})$ and $\mathcal{M}(\boldsymbol{\mathsf{u}})$ with the hyperboloid $\mathcal{Q}(\boldsymbol{\mathsf{u}}) = 0$. In general neither of these level sets, nor the resulting intersections are simple.

We now return to the general constitutive relations (3) and reconsider the pure axial force gedanken experiment close to the unstressed $N = M = 0$ helix $\hat{\boldsymbol{\mathsf{u}}}$, and again pose the question: does a helix over- or under-wind for

$N > 0$ and small? The required argument is in essence the same as the one in the (κ, τ)-plane described in the previous paragraph, but now on the quadric $\mathcal{Q}(\mathbf{u}) = 0$ embedded in three-dimensional Darboux space. The set of possible equilibria under pure axial force are characterized by the intersection of the ellipsoid $\mathcal{E} = 0$ with the quadric $\mathcal{Q} = 0$, which contains the point $\hat{\mathbf{u}}$. Starting from the unstressed configuration and applying a loading of a pure tensile force $N > 0$, a one-parameter family of helices is obtained for small N. In other words the intersection of the two quadrics is a curve $\mathbf{u}_N$ of equilibria in Darboux space parametrized by the force $N \geq 0$ such that $\mathbf{u}_0 = \hat{\mathbf{u}}$. If $|\mathbf{u}_N| > |\hat{\mathbf{u}}|$ close to $\hat{\mathbf{u}}$, then the helix initially overwinds under applied tension and would unwind under applied compression. Geometrically this implies that the loading curve on the hyperboloid with $N > 0$ lies outside the sphere $\mathcal{S}(\mathbf{u}) := \mathbf{u}.\mathbf{u} - \hat{\mathbf{u}}.\hat{\mathbf{u}} = 0$. Therefore, the criterion for deciding whether a given unstressed helix $\hat{\mathbf{u}}$ will overwind under tensile stress is obtained by taking the scalar product of the tangent $\mathbf{t}_{\mathcal{E}}$ to the curve $\mathbf{u}_N$ at $\hat{\mathbf{u}}$ oriented in the direction of increasing N with the exterior normal to the sphere $\mathcal{S}(\mathbf{u})$ at $\hat{\mathbf{u}}$ (see Fig. 3 in Main Text). Specifically, overwinding arises when $\epsilon\hat{\mathbf{u}}.(\mathbf{K}\hat{\mathbf{u}} \times \mathbf{h}) > 0$, where $\mathbf{h} = (-K_1\hat{\mathsf{u}}_2, K_2\hat{\mathsf{u}}_1, K_{23}\hat{\mathsf{u}}_1 - K_{13}\hat{\mathsf{u}}_2)$ is the vector normal to the hyperboloid at $\hat{\mathbf{u}}$ (cf. Fig. 3). Dependent on the details of the constitutive relation, the helix can either overwind or unwind when pulled from the unstressed state. Note also that since the ellipsoid $\mathcal{E} = 0$ is bounded, the family of equilibria $|\mathbf{u}_N|$ will always eventually unwind for large enough N, just as before.

B.COMPUTATION OF THE REDUCED QUADRATIC ENERGY

We show how to compute explicitly the coefficients of the reduced quadratic energy from the displacements of a elastic helical rods under pure wrench. We consider an inextensible, unshearable, uniform rod with a quadratic strain-energy function defined by the positive-definite matrix $\mathbf{K}$ as explained in the text. We assume that the unstressed state of the rod is a helix defined by its Darboux vector $\hat{\mathbf{u}}$. Then, when a wrench (M, N) consisting of an axial torque M and an axial load N is imposed the helical unstressed shape deforms into a new stressed shape defined by a Darboux vector $\mathbf{u}$. For a given Darboux vector $\mathbf{u}$, the shape of the helix is fully determined by two parameters

$$\alpha = |\mathbf{u}|, \qquad \beta = \frac{\mathsf{u}_3}{|\mathbf{u}|}, \tag{5}$$

which are, respectively, the number of turns per unit length and the pitch per unit length (with a sign defining the handedness of the helix, (+) for right-handed helices and arcs of circles, (-) for left-handed helices). The general nonlinear problem consists in finding the shape of the helical rod (α, β) for a given wrench (M, N). To do so, we first define

$$\mathcal{M}(\mathbf{u}) := \epsilon\mathbf{m} \cdot \frac{\mathbf{u}}{|\mathbf{u}|}, \quad \mathcal{N}(\mathbf{u}) := \epsilon\mu|\mathbf{u}|, \quad \mathcal{Q}(\mathbf{u}) := \mathsf{u}_2\mathsf{m}_1 - \mathsf{u}_1\mathsf{m}_2 \tag{6}$$

with $\mu = \mathsf{m}_3 - \mathsf{u}_3(\mathsf{u}_2\mathsf{m}_1 + \mathsf{u}_1\mathsf{m}_2)/(2\mathsf{u}_1\mathsf{u}_2)$ and

$$\mathbf{m} = \mathbf{K}(\mathbf{u} - \hat{\mathbf{u}}), \quad \mathbf{K} = \begin{pmatrix} K_1 & 0 & K_{13} \\ 0 & K_2 & K_{23} \\ K_{13} & K_{23} & K_3 \end{pmatrix}, \ K_1 \leq K_2. \tag{7}$$

For a given (M, N), an helical equilibrium is obtained as a solution of

$$\mathcal{M}(\mathbf{u}) = M, \quad \mathcal{N}(\mathbf{u}) = N, \quad \mathcal{Q}(\mathbf{u}) = 0. \tag{8}$$

As explained in the main text, the condition $\mathcal{Q}(\mathbf{u}) = 0$ implies that (generically) all helical equilibria lie on a single-sheeted hyperboloid in Darboux space. On that hyperboloid, every point is parametrized by two independent parameters such as the pair (α, β). Therefore, for all points on the hyperboloid, we can write $\mathbf{u} = \mathbf{u}(\alpha, \beta)$.

Consider now a particular equilibrium position characterized by the two pairs (M^*, N^*) and (α^*, β^*). The problem is to obtain, close to that point, the effective moduli, that is the coefficient of the 2×2-matrix $\mathbf{G}$, such that

$$\begin{bmatrix} M \\ N \end{bmatrix} - \begin{bmatrix} M^* \\ N^* \end{bmatrix} = \mathbf{G} \begin{bmatrix} \alpha - \alpha^* \\ \beta - \beta^* \end{bmatrix} = \mathbf{G} \cdot \begin{bmatrix} \theta \\ z \end{bmatrix}, \tag{9}$$

where θ and z are, respectively, the rotation angle θ in the plane perpendicular to the axis and the axial extension (as shown in Figure 1 of the main text). That is, we replace the microscopic helical structure by a simpler cylindrical structure that can extend and rotate under torque and tension. For any value of the wrench, the new cylindrical structure is characterised by 3 effective moduli, namely the entries of the symmetric matrix $\mathbf{G}$

$$\mathbf{G} = \begin{bmatrix} A^* & B^* \\ B^* & C^* \end{bmatrix}. \tag{10}$$

In an experiment where the wrench (M, N) is controlled, the corresponding quadratic energy is the following function of the displacements (θ, z),

$$\mathrm{E} = \mathrm{E}^* + (M^* - M)\theta + (N^* - N)z + \frac{1}{2}(A^*\theta^2 + 2B^* z\theta + C^* z^2). \tag{11}$$

Moreover, since we know the variation of M and N away from the reference wrench, the entries of $\mathbf{G}$ can be directly obtained from the functions $(\mathcal{M}, \mathcal{N})$ as

$$\mathbf{G} = \left.\frac{\partial(\mathcal{M}, \mathcal{N})}{\partial(\alpha, \beta)}\right|_{(\alpha=\alpha^*, \beta=\beta^*)}. \tag{12}$$

The variables (α, β) are directly related to the coiling angle and the translation along the axis. However, they are not convenient parametrizations to obtain explicit relations for the moduli. Instead, we use a particular property of helices that allows us to write the moment as [10]

$$\mathbf{m} = a\mathbf{v} + b\mathbf{u}, \quad \mathbf{v} = (0, 0, 1)^{\mathrm{T}}. \tag{13}$$

This relationship, together with (7) can be used to write $\mathbf{u}$ in terms of (u_3, b). Explicitly, we have

$$\mathsf{u}_1 = \frac{K_{13}(\mathsf{u}_3 - \hat{\mathsf{u}}_3) + K_1\hat{\mathsf{u}}_1}{b - K_1}, \tag{14}$$

$$\mathsf{u}_2 = \frac{K_{23}(\mathsf{u}_3 - \hat{\mathsf{u}}_3) + K_2\hat{\mathsf{u}}_2}{b - K_2}, \tag{15}$$

$$\mathsf{u}_3 = \mathsf{u}_3, \tag{16}$$

and axial force and moment can be parametrized in terms of (u_3, b), $\tilde{\mathcal{M}} = \mathcal{M}(\mathbf{u}(\mathsf{u}_3, b)), \tilde{\mathcal{N}} = \mathcal{N}(\mathbf{u}(\mathsf{u}_3, b))$. Given a pair (u_3^*, b^*), we have $M^* = \tilde{\mathcal{M}}(\mathsf{u}_3^*, b^*), N^* = \tilde{\mathcal{M}}(\mathsf{u}_3^*, b^*)$. Similarly, we have $\alpha = \alpha(\mathbf{u}(\mathsf{u}_3, b)), \beta = \beta(\mathbf{u}(\mathsf{u}_3, b))$ so that the Jacobian matrix $\mathbf{J}$ between the two parametrizations is

$$\mathbf{J} = \left.\frac{\partial(\alpha, \beta)}{\partial(\mathsf{u}_3, b)}\right|_{(\mathsf{u}_3=\mathsf{u}_3^*, b=b^*)}. \tag{17}$$

Thanks to this parametrization, it is now a straightforward algebraic computation to compute the moduli

$$\mathbf{G} = \left.\frac{\partial(\tilde{\mathcal{M}}, \tilde{\mathcal{N}})}{\partial(\mathsf{u}_3, b)}\right|_{(\mathsf{u}_3=\mathsf{u}_3^*, b=b^*)} \left.\frac{\partial(\mathsf{u}_3, b)}{\partial(\alpha, \beta)}\right|_{(\mathsf{u}_3=\mathsf{u}_3^*, b=b^*)} = \left.\frac{\partial(\tilde{\mathcal{M}}, \tilde{\mathcal{N}})}{\partial(\mathsf{u}_3, b)}\right|_{(\mathsf{u}_3=\mathsf{u}_3^*, b=b^*)} \mathbf{J}^{-1}. \tag{18}$$

In general the computation of these coefficients is simple, but rather cumbersome, even in the particular case where the equilibrium point is the unstressed shape, expressing the underlying complexity in the relation between helical microstructure and overall cylindrical response of the structure.

As a particularly important example, we consider again the classic case of constitutive relations appropriate for a circular cross-section and with no bend-twist coupling, that is

$$\mathbf{K} = \mathrm{diag}(K_1, K_1, K_3), \ \hat{\mathsf{u}}_1 = 0, \ \hat{\mathsf{u}}_2 > 0. \tag{19}$$

In this case, the computation of the coefficients (A^*, B^*, C^*) is straightforward and leads to

$$A^* = \frac{K_1(\mathsf{u}_2^*)^2 + K_3(\mathsf{u}_3^*)^2}{(\mathsf{u}_2^*)^2 + (\mathsf{u}_3^*)^2}, \tag{20}$$

$$B^* = -\frac{1}{\mathsf{u}_2^*}\left(2K_1\mathsf{u}_2^*\mathsf{u}_3^* - K_1\mathsf{u}_3^*\hat{\mathsf{u}}_2 + K_3\mathsf{u}_2^*\hat{\mathsf{u}}_3 - 2K_3\mathsf{u}_2^*\mathsf{u}_3^*\right), \tag{21}$$

$$C^* = -\frac{1}{(\mathsf{u}_2^*)^3}\left((\mathsf{u}_2^*)^2 + (\mathsf{u}_3^*)^2\right)\left(K_1(\mathsf{u}_2^*)^3 - K_1(\mathsf{u}_2^*)^2\hat{\mathsf{u}}_2 - K_1(\mathsf{u}_3^*)^2\hat{\mathsf{u}}_2 - K_3(\mathsf{u}_2^*)^3\right). \tag{22}$$

A non-monotonic behavior arises when the coefficients B changes sign. This occurs for $(2K_1\mathsf{u}_2^*\mathsf{u}_3^* - K_1\mathsf{u}_3^*\hat{\mathsf{u}}_2 + K_3\mathsf{u}_2^*\hat{\mathsf{u}}_3 - 2K_3\mathsf{u}_2^*\mathsf{u}_3^*) = 0$ and we recover the condition for the inversion of the helical spring under tension given in the main paper (the point **b** in Fig. 2 given by the intersection of the ellipse $M = 0$ and the hyperbola $2(1-\Gamma)\kappa\tau + \Gamma\hat{\tau}\kappa - \hat{\kappa}\tau = 0$).

[1] N. Chouaieb and J. H. Maddocks, J. Elasticity **77**, 221 (2004).