Electronic Supplementary Material (ESI) for Soft Matter.
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Supplementary information for "Electrostatic wrapping of a microfiber around a curved particle"

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Quantifying carboxylic acid groups in the microfibers

To quantitatively characterize the incorporation of carboxylic acid groups in the fibers, we monitored the adsorption of a positively charged dye, toluidene blue O (TBO) into the microfibers, which binds stoichiometrically to carboxylic acid groups.[1] Batches of 1000 microfibers each were suspended in 5 mL of 1.64 mM TBO in 0.1 mM NaOH, where the microfibers adsorbed TBO and exhibited a dark blue color. After mixing the suspension gently on a tube rotator for 15 minutes, the microfibers were removed from the TBO solution and then repeatedly washed and centrifuged in a 0.1 mM NaOH solution until the supernatant was colorless. The TBO-adsorbed fibers were then transferred to 50 vol% acetic acid (in water) to release the TBO from the microfibers. The concentration of TBO released from the fibers was measured with UV-Vis spectroscopy by monitoring the absorbance at 639 nm. The number of carboxyl groups per volume of fiber are presented in Table 1. The measurement error is ±15%.

Table 1: Number of charged groups per volume of microfiber measured from dye adsorption experiments.

Fiber designation	a (μm)	Carboxyl groups per volume of fiber (nm^{-3})
soft	17.5	0.016
soft	20	0.017
soft	22.5	0.018
soft	27.5	0.015
soft	32.5	0.015
rigid	20	0.026

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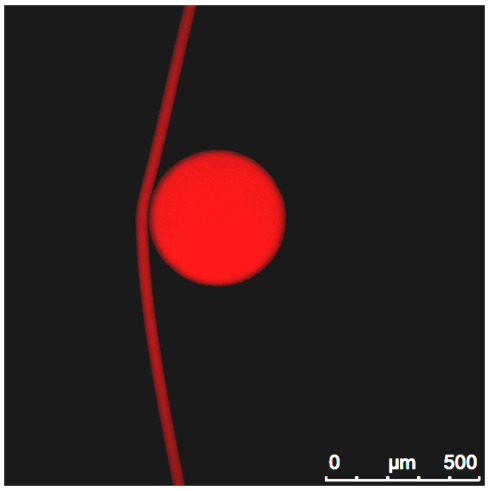

Figure S1: Aggregate formed from a negatively charged fiber, radius $a = 20$ μm, and an uncharged particle, radius $r = 250$ μm and height $= 240$ μm, immersed in 0.1 wt% Tween 80 solution after an external probe pushes the particle into contact with the fiber. The fiber adheres to the particle and remains adhered with gentle agitation of the dish. This experiment verifies that the charged groups on both objects are necessary for wrapping.

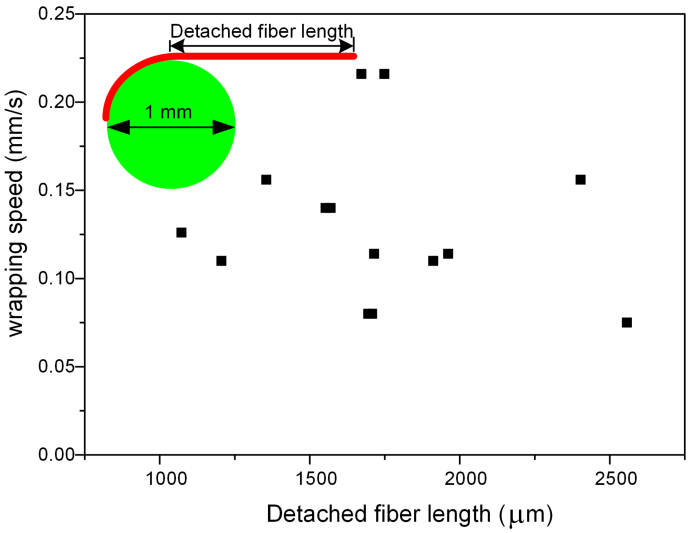

Figure S2: Effect of unattached fiber length on the wrapping rate. Wrapping speed of a negatively charged microfiber, radius $a = 20$ μm, around a positively charged disk-shaped particle, radius $r = 500$ μm and height $= 240$ μm, as a function of the initial unattached fiber length. No systematic dependence on fiber length is observed.

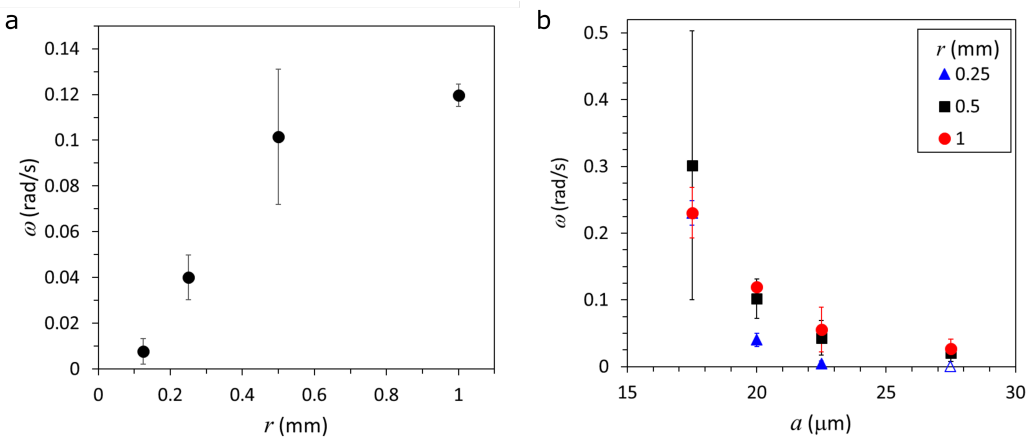

Figure S3: Effect of particle radius r and fiber radius a on the wrapping behavior. (a) Angular rotation (ω = wrapping speed/r) of fiber, $a = 20$ μm, as a function of r. (b) Angular rotation as a function of a for $r = 0.25$, 0.5, 1 mm. Open symbol, where wrapping speed = 0 rad/s, represents adhesion but no wrapping.

Comparison methodology

In this section we outline the methodology to determine the parameter β defined in equation (11) in the manuscript and thus make a prediction for the Young's modulus of the fiber. As discussed in the main text, prior to contact with the particle the fiber has an initial curved prestressed state (see Fig. S4), and so to determine the actual deformation of the fiber during wrapping we must subtract off this original deformation. Our strategy is as follows:

- Calculate the fiber wrapping rate around the particle by reading in coordinates of the point at which the fiber loses contact with the particle at different times. The wrapping rate is found to be constant for a significant proportion of the wrapping, excluding a small start-up transient and the slow down as the fiber attains a steady state. We take the mean wrapping rate for the time over which this is constant.
- Determine the detachment point and thus the detachment angle, α_w, and the attached portion of the fiber, ξ_w.
- Read in the initial prestressed fiber configuration. Parameterize in angle–arclength coordinates as $\theta_p(\xi)$.
- Read in the fiber configuration at a later time, t. Parameterize as $\theta(\xi, t)$. See Fig. S5 for an example.
- Determine the net fiber displacement, ψ using the following relationship:

$$\theta(s,t) = \frac{\pi}{2} - \alpha_w(t) + \theta_p(s) - \theta_p(s_w(t)) + \psi(s,t), \qquad \text{for} \quad s_w(t) \le s \le L. \tag{1}$$

- Solve the system (4)–(7) given in the main text and choose the appropriate value of β such that the L_1 norm between the experimental data and the model prediction is minimized.

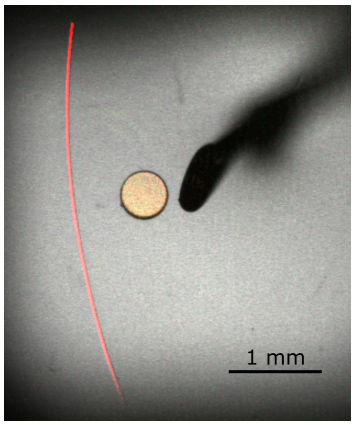

Figure S4: Image of the fiber and particle immersed in 0.1 wt% Tween 80 solution before contact, showing the initial curved (prestressed) profile of the fiber.

- Use this in (13) to predict the Young's modulus.

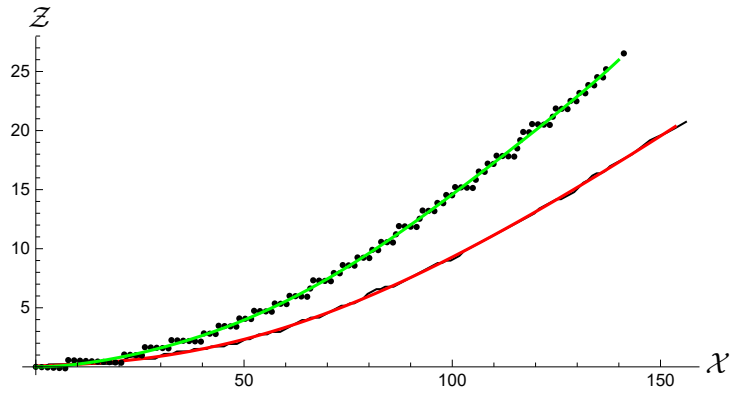

Figure S5: Fits for the prestressed configuration given by $\theta_p(\xi)$ (red) and the deflected profile at a given time t, $\theta(\xi, t)$ (green) for the detached part of the fiber. The raw data is shown in black.

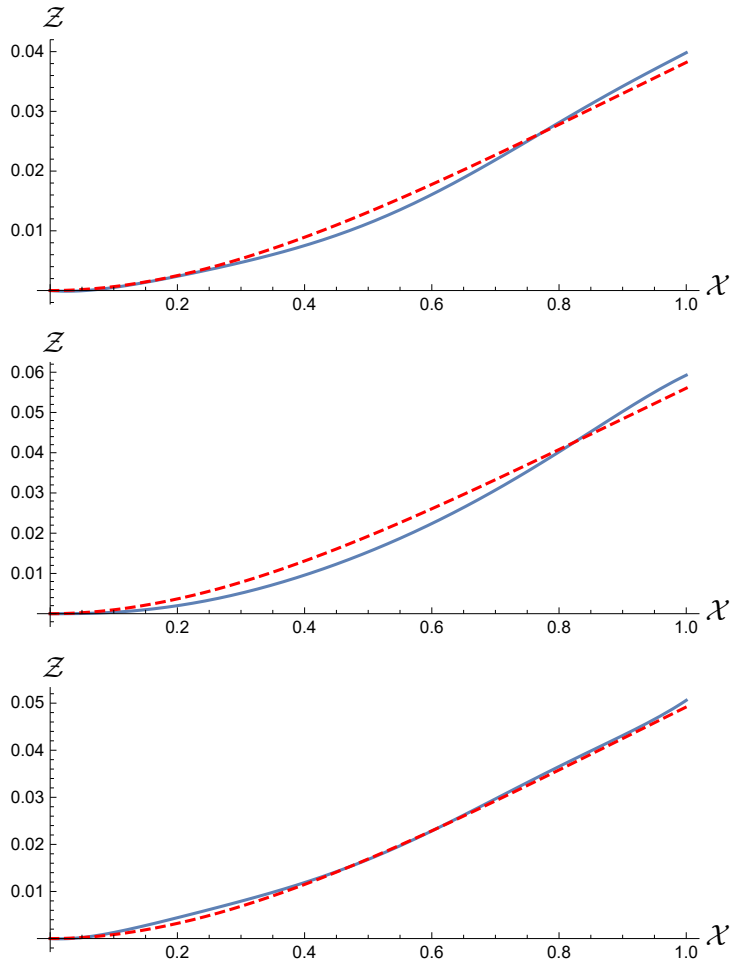

Figure S6: Comparison between theory for the fiber configuration given by the solution (14) in the main text (red dashed) and experimental data (blue solid) for a soft fiber wrapping around a 0.25 mm radius particle in 0.1 mM NaCl. This gives a prediction for the Young's modulus of $E = 71 \pm 6\,\mathrm{kPa}$, where the error corresponds to one standard deviation. We remark that the fit is extremely good and the variability in the prediction between snapshots is low, supporting the reliability of the method.

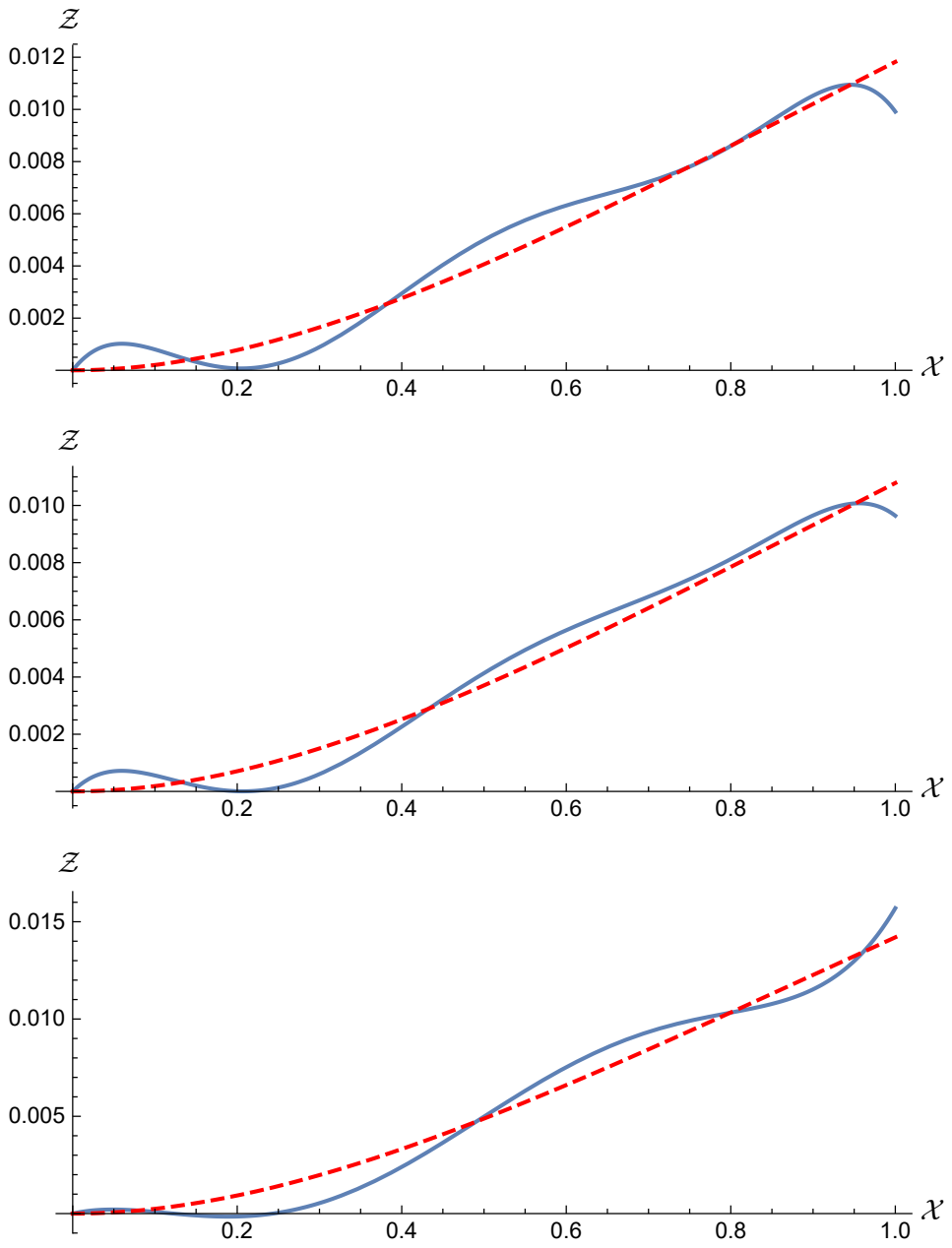

Figure S7: Comparison between theory for the fiber configuration given by the solution to (14) in the main text (red dashed) and experimental data (blue solid) for a rigid fiber wrapping around a 1 mm radius particle in 0.1 mM salt solution. The fit is less good than that of the soft fiber since the images are at the level of the pixel resolution.

References

[1] S. Sano, K. Kato and Y. Ikada, *Biomaterials*, 1993, **14**, 817–822.