Supporting Information

Determination of the Complete Elasticity of *Nephila pilipes* Spider Silk

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Supporting Text

Section S1. Mistakes in two previous BLS studies on the elastic properties of spider silk

The first publication\textsuperscript{43} involves two major mistakes. (i) Three phonon modes were misassigned: the two modes in the “Axial 90a” spectrum (Figure 1a (lower left panel) of ref 43) should correspond to \(L_\parallel\) and \(Q-L\) (or, \(Q-T\) and \(Q-L\), note \(L_\parallel\) and \(Q-T\) have similar frequencies at “Axial 90a”), instead of \(T_\parallel\) and \(L_\parallel\) (or \(T_a\) and \(L_a\), in the terminology of ref 43); the \(T_{r2}\) mode in the “Quasi-radial” spectrum (Figure 1a (lower right panel) of ref 43) should correspond to \(T_\perp\) (or \(T_r1\), in the terminology of ref 43), and thus relate to \(C_{44}\), instead of \(C_{66}\). (ii) In lack of sufficient sound velocities, axial Young’s modulus data of spider silks from stress-strain experiments were used to determine \(C_{13}\), which is inconsistent with the determination of \(C_{11}, C_{12}, C_{33}\), and \(C_{44}\). These two mistakes lead to significant errors in the sound velocities, elastic properties, and final conclusions. The second publication\textsuperscript{44} contains two major mistakes as well. (i) The spider silk was considered as a phononic material, possessing a phononic band gap, which, however, was due to the wrong assignment of a quasi-transverse mode as a longitudinal one and the incorrect presentation of the phonon dispersion relations; in fact, the data points in the so-named branch (3) in Figures 2a,b and 3b of ref 44 result from artificial backscattering phonons and do not belong to the same phonon branch. (ii) The proposed rationalization of the phonon branches was based on two distinct, inconsistent models, instead of a unified model. These two mistakes render the claim of a hypersonic phononic bandgap in spider silk invalid.
Section S2. Transversely isotropic elasticity model

The spider silk was assumed to be transversely isotropic, similar to previous studies.\textsuperscript{41,43} To facilitate the analysis, a “123” coordinate system was constructed, with the “3”-axis parallel to the silk axis. For a transversely isotropic material, the elastic stiffness tensor contains 5 independent elastic constants (here chosen as $C_{11}$, $C_{13}$, $C_{33}$, $C_{44}$, $C_{66}$), and has the following form (in the Voigt notation),

$$
\mathbf{C} = \begin{bmatrix}
C_{11} & C_{11} - 2C_{66} & C_{13} \\
C_{11} - 2C_{66} & C_{11} & C_{13} \\
C_{13} & C_{13} & C_{33} \\
C_{44} & C_{44} & C_{66} \\
\end{bmatrix}.
$$

(S1)

The elastic stiffness constants are coupled with the direction-dependent sound velocities in the framework of the Christoffel’s equation.\textsuperscript{46-49} Given the latter, the former could be uniquely determined through least squares fitting. Because of the transverse isotropy, it is only necessary to consider phonon propagation in a quarter of the “23” plane, i.e., $0^\circ \leq \alpha \leq 90^\circ$ (Figure 1e,f).

For a particular direction represented by an angle, $\alpha$, there exist one quasi-longitudinal (Q-L) mode, one quasi-transverse (Q-T) mode, and one pure-transverse (P-T) mode. The direction-dependent sound velocities of the Q-L, Q-T, and P-T modes can be expressed as follows.

$$
\nu_{Q-L}(\alpha) = \frac{-A_1 + \sqrt{A_1^2 - 4A_2}}{2\rho} \quad \text{(S2)}
$$

$$
\nu_{Q-T}(\alpha) = \frac{-A_1 - \sqrt{A_1^2 - 4A_2}}{2\rho} \quad \text{(S3)}
$$

$$
\nu_{P-T}(\alpha) = \frac{A_3}{\sqrt{\rho}} \quad \text{(S4)}
$$

where,
\[ A_1 = -(\sin^2 \alpha C_{11} + \cos^2 \alpha C_{33} + C_{44}) \]  
(S5)

\[ A_2 = \sin^4 \alpha C_{11} C_{44} + \sin^2 \alpha \cos^2 \alpha (C_{11} C_{33} - C_{13}^2 - 2C_{13} C_{44}) + \cos^4 \alpha C_{33} C_{44} \]  
(S6)

\[ A_3 = \sin^2 \alpha C_{66} + \cos^2 \alpha C_{44}. \]  
(S7)

For simplicity, the Q-L(0°), Q-T(0°), P-T(0°), Q-L(90°), Q-T(90°), P-T(90°), Q-L(0° < \alpha < 90°), Q-T(0° < \alpha < 90°), and P-T(0° < \alpha < 90°) phonon modes are denoted as L\|, T\|, L, T\perp, L\perp, T\perp, L\perp, T\perp, Q-L, Q-T, and P-T, respectively. The density, \( \rho \), of the spider silk was assumed to be 1300 kg m\(^{-3}\), as indicated in previous studies.\textsuperscript{55-57}
Section S3. Brillouin light spectroscopy (BLS) experiments

The direction-dependent sound velocities were obtained by BLS, a non-contact, non-destructive technique. The experiments were conducted with a micro-BLS setup, which includes a six-pass tandem Fabry-Perot interferometer and a Nd/YAG laser ($\lambda_0 = 532$ nm in air) mounted on a goniometer. “Nikon T Plan SLWD 50x” objectives with a numerical aperture (NA) of 0.4 and a super long working distance of 22 mm were used in the measurements to achieve a laser focal spot size $2.0 \pm 0.5$ μm (the $1/e^2$ definition), which is much smaller than the diameter of the silk fiber ($8.3 \pm 0.6$ μm). The laser input power is around 5 mW. Transmission, reflection, and backscattering geometries were employed to probe phonon propagation in directions parallel, normal, and oblique to the spider fiber axis, respectively. In the transmission, reflection, and backscattering geometries, we conducted 4 (scattering angle from 60° to 130°), 5 (scattering angle from 90° to 130°), and 9 (incident angle from 0° to 80°) measurements, respectively. In the transmission and reflection geometries, the direction of the phonon wave vector remains unchanged, but its magnitude varies depending on the scattering angle. In the backscattering geometry, the magnitude of the phonon wave vector slightly changes from 0.00331 to 0.00334 nm$^{-1}$ because the spider silk is slightly birefringent, but its direction varies depending on the laser incident angle. The polarization of the probed phonons (i.e., Q-L, Q-T, P-T) was selected by using different polarization combinations of the incident and scattered light beams (e.g., VV, VH, HH) and meanwhile taking into account the intensity of the scattered light. Considering the optical birefringence of the spider fiber, the magnitudes of the phonon wave vectors probed in the three scattering geometries (e.g., transmission, reflection, backscattering) with the different light polarization configurations (e.g., VV, VH, HH, HV) were calculated as follows.

In the transmission geometry:

$$q_{P, VV} = q_{P, HH} = \frac{4\pi}{\lambda_0} \sin \beta$$  \hspace{1cm} (S8)
\[ q_{p, \text{VH}} = q_{p, \text{HV}} = \frac{2\pi}{\lambda_0} \sqrt{n_0^2 + [n(\beta)]^2 - 2\left(\sqrt{n_0^2 - \sin^2 \beta} \sqrt{[n(\beta)]^2 - \sin^2 \beta - \sin^2 \beta}\right)} \]  

(S9)

In the reflection geometry:

\[ q_{\perp, \text{VV}} = \frac{4\pi}{\lambda_0} \sqrt{n_0^2 - \sin^2 \beta} \]  

(S10)

In the backscattering geometry:

\[ q_{\text{bs, VV}} = \frac{4\pi n_0}{\lambda_0} \]  

(S11)

\[ q_{\text{bs, HH}} = \frac{4\pi n(\beta)}{\lambda_0} \]  

(S12)

\[ q_{\text{bs, VH}} = q_{\text{bs, HV}} = \frac{2\pi}{\lambda_0} \sqrt{n_0^2 + [n(\beta)]^2 + 2\left(\sqrt{n_0^2 - \sin^2 \beta} \sqrt{[n(\beta)]^2 - \sin^2 \beta + \sin^2 \beta}\right)} \]

(S13)

Here, \( \beta \) is the laser incident angle, and \( n \) is the spider fiber’s refractive index. It should be noted that the optical birefringence affects both the magnitude, \( q \), and direction, \( \alpha \), of the phonon wave vector, and that \( \alpha \) is related to \( \beta \) by the Snell’s law, \( \sin \beta = n \sin \gamma \), where \( \gamma = 90^\circ - \alpha \) (Figures 1e and S1). The sound velocity was calculated as,

\[ v = \frac{2\pi f}{q}, \]

(S14)

where \( f \) is the phonon frequency obtained from the Lorentzian fits to the Brillouin peaks. In the experiments studying the strain effect on the spider silk’s elastic properties, the silk fiber was stretched using a customized stretch meter by up to 20%. 

6
Section S4. Optical birefringence

The spider silk was found to be optically birefringent from polarized optical microscopy measurements (inset to Figure S2). To account for the light-polarization-dependent refractive index, the following formula was adopted,

\[ n(\phi) = \left( \frac{\sin^2 \phi}{n_o^2} + \frac{\cos^2 \phi}{n_e^2} \right)^{-1/2}, \quad (S15) \]

where \( \phi \) is the angle between the light polarization vector and the fiber axis. A “V”-polarized light beam has a refractive index of \( n_V = n_o \), because \( \phi = 90^\circ \), whereas \( n_H = n(\beta) \) for an “H”-polarized light beam.

Therefore, in the backscattering measurements with \( \beta = 0^\circ \),

\[ f_{\text{VV}} = \frac{v_L}{2\pi} q = \frac{v_L}{2\pi} \frac{4\pi n}{\lambda_0} = \frac{2n_o v_L}{\lambda_0} \quad (S16) \]

\[ f_{\text{HH}} = \frac{2n_e v_L}{\lambda_0}, \quad (S17) \]

where \( v_L \) is the sound velocity of the \( L_\perp \) mode. The frequency shift in Figure S2 indicates the optical anisotropy, i.e., \( \frac{n_o}{n_e} = \frac{f_{\text{VV}}}{f_{\text{HH}}} \). Based on eqs S10 and S14, the \( v_L \) and \( n_o \) were obtained by conducting BLS measurements in the reflection geometry at multiple \( \beta \) values. Finally, the principal refractive indices were determined to be \( n_o = 1.40 \pm 0.01 \) and \( n_e = 1.46 \pm 0.02 \).
Section S5. \( \chi^2 \) fitting

Based on the BLS-measured, direction-dependent sound velocities (i.e., \( v_{Q-L}(\alpha), v_{Q-T}(\alpha), v_{P-T}(\alpha) \)), nonlinear \( \chi^2 \) fitting was conducted to obtain the elastic stiffness constants.\(^{50}\) The \( \chi^2 \) is defined as

\[
\chi^2 = \sum_i \left[ \frac{v_{i,\text{fit}}(C_{11}, C_{13}, C_{33}, C_{44}, C_{66}, \alpha) - v_{i,\text{exp}}(\alpha)}{(\Delta v_{i,\text{exp}})^2} \right]^2,
\]

where \( v_{i,\text{fit}} \) and \( v_{i,\text{exp}} \) are the fitted and experimental sound velocities, respectively, \( \Delta v_{i,\text{exp}} \) is the uncertainty of the measured sound velocity, and the summation is over all experimental sound velocities. By considering the measurement uncertainties of the angles, refractive indices, phonon frequencies, and so on, the relative uncertainty of \( (\Delta v_{i,\text{exp}})/(v_{i,\text{exp}}) \) was estimated to be 2%. To ensure positive Young’s and shear moduli, the \( \chi^2 \) fitting was performed subject to the following constraints: (i) \( C_{11} > |C_{12}| \), (ii) \( C_{44} > 0 \), and (iii) \( C_{33}(C_{11} + C_{12}) > 2C_{13}^2 \).\(^{52}\) Note \( C_{12} = C_{11} - 2C_{66} \). The fitting was based on 8-10 data points for the Q-L modes, 3-6 data points for the Q-T modes, and 0-3 data points for the P-T modes. The final \( \chi^2 \) values are 8.7073, 0.7172, 1.0634, 0.6167, and 1.5541, for the data sets corresponding to 0%, 5%, 10%, 15%, and 20% strains, respectively. These \( \chi^2 \) values indicate that qualitatively the discrepancy between the experimental and predicted sound velocities is within \( (8.7073)^{0.5}\Delta v_{\text{exp}} = 2.95\Delta v_{\text{exp}} \). Since \( \Delta v_{\text{exp}} \approx 2\% v_{\text{exp}} \), the discrepancy is then within 5.9% (standard deviation). The discrepancy for the other four data sets is even smaller (within 2.5%). The residuals for the 0% strain data set, which has the largest \( \chi^2 \) value, are shown in Figure S5. Similar residual profiles exist for the other four data sets. In Figure S5, no clear pattern of the residuals is seen. Therefore, we consider the residuals to be randomly distributed. At 0% strain, the availability of the \( v_{Q-L}(\alpha), v_{Q-T}(\alpha), \) and \( v_{P-T}(\alpha) \) data at multiple \( \alpha \) values allows unique determination of \( C_{11}, C_{13}, C_{33}, C_{44}, \) and \( C_{66} \). At higher strains, where no unambiguous P-T modes were detected, the determination of the elastic tensors was completed by assuming the Poisson’s ratio \( v_{31} \) to be equal to that at 0% strain (i.e.,
\( \nu_{31} = 0.35 \pm 0.02 \). The obtained elastic stiffness constants of the spider fiber at the five strains are summarized in Table S1.

The independent elastic stiffness constants were used to predict the theoretical \( v_{Q\perp}(\alpha) \), \( v_{Q\parallel}(\alpha) \), and \( v_{P\parallel}(\alpha) \), according to eqs S2-S4. Furthermore, they were used to calculate the characteristic mechanical properties,\(^{41}\) including the axial and lateral Young’s moduli (\( E_{\parallel}, E_{\perp} \)), shear moduli (\( G_{13} = G_{23}, G_{12} \)), and Poisson’s ratios (\( \nu_{31} = \nu_{32}, \nu_{12} \)). Note that only five of the engineering mechanical properties are independent. Typically, \( \{ E_{\parallel}, E_{\perp}, G_{13}, G_{12}, \nu_{31} \} \) or \( \{ E_{\parallel}, E_{\perp}, G_{13}, \nu_{12}, \nu_{31} \} \) are chosen. The relevant formulas and physical meanings of the mechanical properties are summarized in Table S2. Their values for the *Nephila pilipes* MA silk in the native and stretched states are summarized in Table S3.
Section S6. Uncertainty quantification

By considering the measurement uncertainties of the angles, refractive indices, phonon frequencies, and so on, the relative uncertainty of the BLS-measured sound velocities, $(Δv_i, \text{exp})/(v_i, \text{exp})$, was estimated to be 2%. The uncertainties of the elastic stiffness constants were determined by constructing a matrix $G$. To simplify the notations, $C_{11}$, $C_{13}$, $C_{33}$, $C_{44}$, and $C_{66}$ were denoted as $a_1$, $a_2$, $a_3$, $a_4$, and $a_5$, respectively. $G$ has elements of the following form,

$$G_{mn} = \sum_i \frac{1}{(Δv_{i, \text{exp}})^2} \frac{∂v_{i, \text{fit}}}{∂a_m} \frac{∂v_{i, \text{fit}}}{∂a_n}, \quad (S19)$$

where $m$ and $n (= 1, 2, 3)$ are matrix indices, and the summation is over all experimental sound velocities. Mathematically, $G$ dictates how variations in the elastic constants affect the fitted sound velocities. The partial derivatives were approximated by using a central finite difference scheme. For example,

$$\frac{∂v_{i, \text{fit}}}{∂a_m} = \frac{v_{i, \text{fit}}(a_m + δa_m) - v_{i, \text{fit}}(a_m - δa_m)}{2δa_m},$$

where $δa_m$ represents the change in $a_m$. From a convergence study, $δa_m$ was determined to be $10^{-6}a_m$. The inverse of $G$ was calculated to obtain a covariance matrix, i.e., $M = G^{-1}$. The uncertainty of $a_m$ is then the square root of the $m^{th}$ diagonal term of $M$ (i.e., $Δa_m = √{M_{mm}}$). As an example, the $M$ matrix for the spider silk at 0% strain is shown below.

$$M(\text{unit: } 10^{18} \text{ (GPa)}^2) = \begin{bmatrix}
0.08490 & 0.02809 & -0.08048 & -0.00744 & 0.00250 \\
0.02809 & 0.08582 & 0.11208 & -0.01213 & 0.00408 \\
-0.08048 & 0.11208 & 0.60955 & -0.03251 & 0.01094 \\
-0.00744 & -0.01213 & -0.03251 & 0.02241 & -0.00754 \\
0.00250 & 0.00408 & 0.01094 & -0.00754 & 0.01855
\end{bmatrix}. \quad (S20)$$

The uncertainties of the Young’s moduli, shear moduli, and Poisson’s ratios were calculated according to the principles of uncertainty propagation (i.e., the chain rule). For example,
\[ \Delta E_p = \sqrt{\sum_{j, k=11, 13, 33, 66} \left( \frac{\partial E_p}{\partial C_{ij}} \right)^2 + \sum_{j, k=11, 13, 33, 66} 2 \frac{\partial E_p}{\partial C_{ij}} \frac{\partial E_p}{\partial C_{kl}} (\Delta C_{ij})(\Delta C_{kl})}, \quad (S21) \]

where \( \frac{\partial E_p}{\partial C_{11}} = \frac{C_{13}^2}{(C_{11} - C_{66})^2} \), \( \frac{\partial E_p}{\partial C_{13}} = -\frac{2C_{13}}{C_{11} - C_{66}} \), \( \frac{\partial E_p}{\partial C_{33}} = 1 \), and \( \frac{\partial E_p}{\partial C_{66}} = -\frac{C_{13}^2}{(C_{11} - C_{66})^2} \). Here, \((\Delta C_{ij})(\Delta C_{kl})\) represents the covariance of \( C_{ij} \) and \( C_{kl} \). \((\Delta C_{ij})(\Delta C_{kl})\) are the off-diagonal terms of the matrix \( M \). For example, \((\Delta C_{11})(\Delta C_{13}) = M(1, 2)\), \((\Delta C_{11})(\Delta C_{33}) = M(1, 3)\), \((\Delta C_{11})(\Delta C_{66}) = M(1, 5)\), \((\Delta C_{13})(\Delta C_{33}) = M(2, 3)\), \((\Delta C_{13})(\Delta C_{66}) = M(2, 5)\), and \((\Delta C_{33})(\Delta C_{66}) = M(3, 5)\). Similar expressions for \( E_{\perp}, G_{13}, G_{12}, \nu_{31}, \) and \( \nu_{12} \) are shown below.

\[ \Delta E_{\perp} = \sqrt{\sum_{j, k=11, 13, 33, 66} \left( \frac{\partial E_{\perp}}{\partial C_{ij}} \right)^2 + \sum_{j, k=11, 13, 33, 66} 2 \frac{\partial E_{\perp}}{\partial C_{ij}} \frac{\partial E_{\perp}}{\partial C_{kl}} (\Delta C_{ij})(\Delta C_{kl})}, \quad (S22) \]

where \( \frac{\partial E_{\perp}}{\partial C_{11}} = \frac{4C_{13}^2 C_{66}}{(C_{11} - C_{33} - C_{13})^2} \), \( \frac{\partial E_{\perp}}{\partial C_{13}} = -\frac{8C_{13} C_{33} C_{66}}{(C_{11} - C_{33} - C_{13})^2} \), \( \frac{\partial E_{\perp}}{\partial C_{33}} = \frac{4C_{13}^2 C_{66}}{(C_{11} - C_{33} - C_{13})^2} \), and \( \frac{\partial E_{\perp}}{\partial C_{66}} = 4 - \frac{8C_{13} C_{66}}{C_{11} - C_{33} - C_{13}} \).

\[ \Delta G_{13} = \Delta G_{23} = \Delta C_{44}. \quad (S23) \]

\[ \Delta G_{12} = \Delta C_{66}, \quad (S24) \]

\[ \Delta \nu_{31} = \sqrt{\sum_{j, k=11, 13, 33, 66} \left( \frac{\partial \nu_{31}}{\partial C_{ij}} \right)^2 + \sum_{j, k=11, 13, 33, 66} 2 \frac{\partial \nu_{31}}{\partial C_{ij}} \frac{\partial \nu_{31}}{\partial C_{kl}} (\Delta C_{ij})(\Delta C_{kl})}, \quad (S25) \]

where \( \frac{\partial \nu_{31}}{\partial C_{11}} = -\frac{C_{13}}{2(C_{11} - C_{66})} \), \( \frac{\partial \nu_{31}}{\partial C_{13}} = \frac{1}{2(C_{11} - C_{66})} \), \( \frac{\partial \nu_{31}}{\partial C_{33}} = 0 \), and \( \frac{\partial \nu_{31}}{\partial C_{66}} = \frac{C_{13}}{2(C_{11} - C_{66})} \).

\[ \Delta \nu_{12} = \sqrt{\sum_{j, k=11, 13, 33, 66} \left( \frac{\partial \nu_{12}}{\partial C_{ij}} \right)^2 + \sum_{j, k=11, 13, 33, 66} 2 \frac{\partial \nu_{12}}{\partial C_{ij}} \frac{\partial \nu_{12}}{\partial C_{kl}} (\Delta C_{ij})(\Delta C_{kl})}, \quad (S26) \]
where \[
\frac{\partial \nu_{12}}{\partial C_{11}} = \frac{2C_{33}^2 C_{66}}{(C_{11} C_{33} - C_{13}^2)^2}, \quad \frac{\partial \nu_{12}}{\partial C_{13}} = \frac{-4C_{13} C_{33} C_{66}}{(C_{11} C_{33} - C_{13}^2)^2}, \quad \frac{\partial \nu_{12}}{\partial C_{33}} = \frac{2C_{13}^2 C_{66}}{(C_{11} C_{33} - C_{13}^2)^2}, \quad \text{and}
\]
\[
\frac{\partial \nu_{12}}{\partial C_{33}} = \frac{-2C_{33}}{C_{11} C_{33} - C_{13}^2}.
\]
Section S7. Scattering intensity calculations

The appearance/disappearance of the phonon modes in a BLS spectrum is determined by the light scattering selection rules. To understand the Pockels (elasto-optic) coefficients, $P_{ij}$, involved in the different measurements, the polarization vectors of the scattered light corresponding to the phonon modes in the transmission, reflection, and backscattering measurements were derived, according to the theory by Hamaguchi, as summarized in the Table S4. The intensity of the scattered light is proportional to $|\xi_\mu|^2/\nu_\mu^2$, where $\xi$ is the polarization vector of the scattered light by a phonon mode $\mu$. Note $\mu = L||, T_{||,1}, T_{||,2}, L\perp, T\perp,1, T\perp,2, Q-L, Q-T, \text{or} P-T$. The $\xi$ expressions are useful for analyzing the appearance/disappearance of the phonon modes in BLS spectra. For example, the disappearance of the $T_{||,2}$ mode in the transmission VH or HV measurements could be attributed to the vanishing $P_{44}$; similar conclusions could be drawn for the reflection and backscattering measurements. In addition, the $T_{||,1}$ and $T\perp,1$ modes are non-detectable in the transmission and reflection VV measurements, respectively, because the corresponding intensities of the scattered light are exactly zero.

It is worth considering the P-T mode in the backscattering measurements. This mode remained non-detectable for the stretched spider fibers. Since the spider fiber is only weakly birefringent (Figure S2), it is reasonable to assume $\gamma_i = 90^\circ - \alpha$ and $\gamma_i = 90^\circ - \alpha$ in the backscattering VH and HV measurements, respectively. Therefore, the invisibility of the P-T mode implies

$$\frac{P_{11} - P_{22}}{2} = P_{44} \frac{n_i^2}{n_0^2}. $$

Analogizing from $P_{ij}$ to $C_{ij}$ leads to$$\frac{C_{11} - C_{12}}{2} = C_{44} \frac{n_i^2}{n_0^2}. $$

As a result,

$$C_{66} = n_0^2 n_i^2 \frac{C_{44}}{1.46^2} = 1.40^2 \frac{C_{44}}{1.40^2} = 1.09C_{44}, $$

which is in good agreement with the data for the stretched spider fibers in Table S1.
Supporting Figures

Figure S1. Schematic of the transmission geometry for BLS experiments. The gray rectangle represents a spider fiber, with a “123” coordination system attached to it. The “3”-axis is parallel to the fiber axis. The green triangle and blue rectangle represent the laser source and Fabry-Perot interferometer, respectively. $\beta$ and $\gamma$ are the laser incident angles outside and inside of the spider fiber, respectively. $k_i$, $k_s$, and $k_{bs}$ are the wave vectors of the incident, scattered, and back-scattered light inside the fiber, respectively. $q_{\parallel}$ and $q_{bs}$ are the phonon wave vectors in the transmission and backscattering geometries, respectively. The internally reflected light leads to the artificial backscattering modes in the transmission spectra, as seen in Figure 2a. $n$ represents the refractive index of the spider fiber.
Figure S2. VV and HH BLS spectra at $\beta = 0^\circ$ in the backscattering geometry. The experimental data (gray lines) are represented by Lorentzian shapes (red lines). The dashed line indicates the frequency shift between the VV and HH spectra. Inset: polarized optical microscopy image of the spider fiber.
**Figure S3.** (a) Representative experimental HH BLS spectra including both the Stokes and anti-Stokes sides recorded in the backscattering geometry at incident angles, $\beta = 30^\circ$, $40^\circ$, and $50^\circ$. 

![Graph showing HH BLS spectra with different incident angles](image)
Figure S4. (a) Comparison of VV BLS spectra recorded in the transmission (black) and backscattering (blue) geometries at the same incident angle, $\beta = 45^\circ$. The anti-Stokes side of transmission spectrum is well represented by the Lorentzian fit (red line) with the individual contributions shown by blue lines, allowing correct assignments of the phonon modes. The dash lines on the Stokes side denote the positions of the Q-T and Q-L modes resolved in the backscattering geometry. (b) The frequencies of the Q-L and Q-T modes in the transmission and backscattering spectra as a function of the incident angle.
Figure S5. Residuals of the fitted sound velocities for the *Nephila pilipes* MA spider silk at 0% strain. Similar residual profiles exist for the data at 5%, 10%, 15%, and 20% strains.
Figure S6. VH backscattering spectrum recorded at an incident angle $\beta = 40^\circ$. The anti-Stokes side is well-represented by the Lorentzian fit (red line) with the individual contributions shown by blue lines.
Figure S7. Angle-dependent sound velocities in the stretched spider fiber at (a) 5%, (b) 10%, and (c) 15% strains. \( \alpha \) represents the angle between the phonon wave vector and the fiber axis. Q-L, Q-T, and P-T denote the quasi-longitudinal, quasi-transverse, and pure-transverse phonon modes, respectively. The lines represent theoretical predictions by eqs S2-S4. Error bars smaller than the symbol size are not shown.
Supporting Tables

**Table S1.** Summary of the elastic stiffness constants of the *Nephila pilipes* MA silk at five strains (i.e., 0%, 5%, 10%, 15%, 20%). The zero and positive strains indicate spider fibers in the native and stretched states, respectively.

| Strain | $C_{11}$ (GPa) | $C_{13}$ (GPa) | $C_{33}$ (GPa) | $C_{44}$ (GPa) | $C_{66}$ (GPa) | $C_{12}$ (GPa) |
|--------|----------------|----------------|----------------|----------------|----------------|----------------|
| 0%     | 14.4 ± 0.3     | 8.0 ± 0.3      | 26.6 ± 0.8     | 4.1 ± 0.1      | 3.1 ± 0.1      | 8.2 ± 0.4      |
| 5%     | 14.0 ± 0.3     | 7.0 ± 0.5      | 28.8 ± 1.1     | 3.9 ± 0.5      | 4.0 ± 0.9      | 6.0 ± 1.9      |
| 10%    | 13.8 ± 0.3     | 6.9 ± 0.5      | 30.3 ± 1.2     | 4.2 ± 0.5      | 3.9 ± 1.0      | 5.9 ± 1.9      |
| 15%    | 13.7 ± 0.3     | 6.6 ± 0.5      | 31.2 ± 1.2     | 4.1 ± 0.6      | 4.3 ± 1.0      | 5.2 ± 1.9      |
| 20%    | 13.7 ± 0.3     | 6.9 ± 0.5      | 32.4 ± 1.2     | 3.6 ± 0.6      | 3.8 ± 0.9      | 6.0 ± 1.9      |
Table S2. Summary of the definitions, formulas, and physical meanings of the axial and lateral Young’s moduli, shear moduli, and Poisson’s ratios considered in this work. The subscripts (“1”, “2”, “3”) refer to the “123” coordinate system shown in Figure 1f. The formulas are adapted from ref 41. Note that ν₃₁ was mistakenly denoted as ν₁₃ in ref 41.

| Quantity (unit) | Formula | Physical meaning |
|-----------------|---------|------------------|
| Axial Young’s modulus (GPa) | \( E_p = C_{33} - \frac{C_{13}^2}{C_{11} - C_{66}} \) | Resistance to uniaxial stress parallel to “3”-axis |
| Lateral Young’s modulus (GPa) | \( E_\perp = \frac{4C_{66} \left[ C_{33}(C_{11} - C_{66}) - C_{13}^2 \right]}{C_{11}C_{33} - C_{13}^2} \) | Resistance to uniaxial stress normal to “3”-axis |
| Shear modulus in “1-3” or “2-3” plane (GPa) | \( G_{13} = G_{23} = C_{44} \) | Resistance to shear stress on a plane normal to “3”-axis and along “1”- or “2”-axis |
| Shear modulus in “1-2” plane (GPa) | \( G_{12} = C_{66} \) | Resistance to shear stress on a plane normal to “2”-axis and along “1”-axis |
| Poisson’s ratio in “1-3” or “2-3” plane | \( \nu_{31} = \nu_{32} = \frac{C_{13}}{2(C_{11} - C_{66})} \) | Response of strain (\( \varepsilon \)) in direction “1” or “2” due to a strain in direction “3”; \( \nu_{31} = \frac{\varepsilon_{11}}{\varepsilon_{33}}; \nu_{32} = \frac{\varepsilon_{22}}{\varepsilon_{33}}. \) |
| Poisson’s ratio in “1-2” plane | \( \nu_{12} = \frac{(C_{11} - 2C_{66})C_{33} - C_{13}^2}{C_{11}C_{33} - C_{13}^2} \) | Response of strain in direction “2” due to a strain in direction “1”; \( \nu_{12} = \frac{\varepsilon_{22}}{\varepsilon_{11}}. \) |
Table S3. Summary of the axial and lateral Young’s moduli, shear moduli, and Poisson’s ratios of the *Nephila pilipes* MA silk at five strains (i.e., 0%, 5%, 10%, 15%, 20%).

| Strain | $E_\parallel$ (GPa) | $E_\perp$ (GPa) | $E_\parallel/E_\perp$ | $G_{13/23}$ (GPa) | $G_{12}$ (GPa) | $\nu_{31/32}$ | $\nu_{12}$ |
|--------|---------------------|-----------------|----------------------|-------------------|---------------|--------------|-----------|
| 0%     | 20.9 ± 0.8          | 9.2 ± 0.3       | 2.3                  | 4.1 ± 0.1         | 3.1 ± 0.1     | 0.35 ± 0.02  | 0.48 ± 0.03|
| 5%     | 23.9 ± 1.2          | 10.8 ± 1.4      | 2.2                  | 3.9 ± 0.5         | 4.0 ± 0.9     | 0.35 ± 0.04  | 0.35 ± 0.15|
| 10%    | 25.5 ± 1.2          | 10.7 ± 1.4      | 2.4                  | 4.2 ± 0.5         | 3.9 ± 1.0     | 0.35 ± 0.04  | 0.36 ± 0.16|
| 15%    | 26.6 ± 1.3          | 11.2 ± 1.3      | 2.4                  | 4.1 ± 0.6         | 4.3 ± 1.0     | 0.35 ± 0.04  | 0.31 ± 0.16|
| 20%    | 27.6 ± 1.3          | 10.5 ± 1.5      | 2.6                  | 3.6 ± 0.6         | 3.8 ± 0.9     | 0.35 ± 0.04  | 0.37 ± 0.15|
Table S4. Summary of the polarization vectors of the scattered light corresponding to the \{L_{\parallel}, T_{\parallel,1}, T_{\parallel,2}\} modes in transmission measurements, \{L_{\perp}, T_{\perp,1}, T_{\perp,2}\} modes in reflection measurements, and \{Q-L, Q-T, P-T\} modes in backscattering measurements. The polarization vectors determine the detectability of the phonon modes. The expressions in this table are adapted from ref S3 and applicable to transversely isotropic materials that are optically birefringent. $P_{ij}$: Pockels (elasto-optic) coefficients ($i, j = 1, 2, 3$).

| BLS geometry | Expressions |
|--------------|-------------|
| **Transmission** | | 
| L\_1 mode: detectable in VV and HH, but not in VH or HV | \[ \xi_{\text{VV}}^{L_1} = [0, -P_{13}, 0] \]
| | \[ \xi_{\text{HH}}^{L_1} = \begin{bmatrix} -P_{33}n_e^4 \cos \alpha \sin^2 \alpha + P_{13}n_o^4 \cos^3 \alpha \n_o^4 \\ 0, -P_{33}n_e^4 \sin^3 \alpha + P_{13}n_o^4 \cos^3 \alpha \sin \alpha \n_o^4 \end{bmatrix} \] |
| T\_\parallel,1 mode: non-detectable | \[ \xi_{\text{VV}}^{T_{\parallel,1}} = \xi_{\text{HH}}^{T_{\parallel,1}} = \xi_{\text{IH}}^{T_{\parallel,1}} = [0, 0, 0] \] |
| T\_\parallel,2 mode: detectable in VH and HV, but not in VV or HH | \[ \xi_{\text{HI}}^{T_{\parallel,2}} = \begin{bmatrix} -\cos \gamma_s \sin \gamma_s P_{44} n_e^2 / n_o^2 \n_o^2, 0, -\cos^2 \gamma_s P_{44} n_e^2 / n_o^2 \n_o^2 \end{bmatrix} \]
| | \[ \xi_{\text{HV}}^{T_{\parallel,2}} = \begin{bmatrix} 0, -P_{44} n_e^2 \cos \gamma_s \n_o^2, 0 \end{bmatrix} \] |
| **Reflection** | | 
| L\_\perp mode: detectable in VV and HH, but not in VH or HV | \[ \xi_{\text{VV}}^{L_{\perp}} = [0, -P_{12}, 0] \]
| | \[ \xi_{\text{HH}}^{L_{\perp}} = \begin{bmatrix} -P_{33}n_e^4 \cos \alpha \sin^2 \alpha + P_{12}n_o^4 \cos^3 \alpha \n_o^4 \\ 0, -P_{33}n_e^4 \sin^3 \alpha + P_{12}n_o^4 \cos^3 \alpha \sin \alpha \n_o^4 \end{bmatrix} \] |
| T\_\perp,1 mode: non-detectable | \[ \xi_{\text{VV}}^{T_{\perp,1}} = \xi_{\text{HH}}^{T_{\perp,1}} = \xi_{\text{TI}}^{T_{\perp,1}} = [0, 0, 0] \] |
| T\_\perp,2 mode: detectable in VH and HV, but not in VV or HH | \[ \xi_{\text{VI}}^{T_{\perp,2}} = \begin{bmatrix} -\sin^2 \gamma_s (P_{11} - P_{12}) / 2, 0, \cos \gamma_s \sin \gamma_s (P_{11} - P_{12}) / 2 \end{bmatrix} \]
| | \[ \xi_{\text{HV}}^{T_{\perp,2}} = \begin{bmatrix} 0, (P_{11} - P_{12}) \sin \gamma_s, 0 \end{bmatrix} \] |
Backscattering

### Q-L mode: detectable in VV and HH, but not in VH or HV

\[
\varepsilon_{VV}^{QL} = \left[ 0, -\left( P_{12} \sin^2 \alpha + P_{13} \cos^2 \alpha \right), 0 \right]
\]

\[
\varepsilon_{HH}^{QL} = \begin{bmatrix}
P_{31} \frac{n_e^4}{n_o^4} \cos \alpha \sin^4 \alpha + \left( -4 P_{44} \frac{n_e^2}{n_o^2} + P_{33} \frac{n_e^4}{n_o^4} + P_{11} \right)^3 \cos \alpha \sin^2 \alpha + P_{13} \cos^5 \alpha \\
0 \\
-P_{13} \cos^4 \alpha \sin \alpha + \left( 4 P_{44} \frac{n_e^2}{n_o^2} - P_{33} \frac{n_e^4}{n_o^4} - P_{11} \right)^2 \cos \alpha \sin^3 \alpha - P_{31} \frac{n_e^4}{n_o^4} \sin^5 \alpha
\end{bmatrix}
\]

### Q-T mode: detectable in VV and HH, but not in VH or HV

\[
\varepsilon_{VV}^{QT} = \left[ 0, -(P_{12} - P_{13}) \cos \alpha \sin \alpha, 0 \right]
\]

\[
\varepsilon_{HH}^{QT} = \begin{bmatrix}
-2 P_{44} \frac{n_e^2}{n_o^2} \left( \cos^2 \alpha - \sin^2 \alpha \right) \cos \alpha \sin \alpha + (P_{11} - P_{13}) \frac{n_e^4}{n_o^4} \cos^3 \alpha \sin^2 \alpha + (P_{11} - P_{13}) \sin \alpha \cos^4 \alpha \\
0 \\
2 P_{44} \frac{n_e^2}{n_o^2} \left( \cos^2 \alpha - \sin^2 \alpha \right) \cos \alpha \sin^2 \alpha - (P_{11} - P_{13}) \frac{n_e^4}{n_o^4} \cos \alpha \sin^4 \alpha - (P_{11} - P_{13}) \cos^3 \alpha \sin^2 \alpha
\end{bmatrix}
\]

### P-T mode: detectable in VH and HV, but not in VV or HH

\[
\varepsilon_{VV}^{PT} = \begin{bmatrix}
\cos \gamma_s \sin \gamma_s P_{44} \frac{n_e^2}{n_o^2} \cos \alpha - \sin^2 \gamma_s \frac{P_{11} - P_{12}}{2} \sin \alpha \\
0 \\
-\cos^2 \gamma_s P_{44} \frac{n_e^2}{n_o^2} \cos \alpha + \cos \gamma_s \sin \gamma_s \frac{P_{11} - P_{12}}{2} \sin \alpha
\end{bmatrix}
\]

\[
\varepsilon_{HH}^{PT} = \left[ 0, -P_{44} \frac{n_e^2}{n_o^2} \cos \alpha \cos \gamma_s + \left( \frac{P_{11} - P_{12}}{2} \right) \sin \alpha \sin \gamma_i, 0 \right]
\]

### Notes

1. \( \gamma_s \) and \( \gamma_i \) can be determined from \( n(\beta) \sin \gamma_s = \sin \beta \) and \( n(\beta) \sin \gamma_i = \sin \beta \), respectively, where \( n(\beta) = \left( \frac{\cos^2 \beta}{n_e^2} + \frac{\sin^2 \beta}{n_o^2} \right)^{-1/2} \) (see eq S15). For weakly birefringent materials, \( \gamma_s \approx 90^\circ - \alpha \) for VH measurements, and \( \gamma_i \approx 90^\circ - \alpha \) for HV measurements.
2. See Figure S1 for the definitions of \( \beta \) and \( \gamma_s \); see Figure 1e for the definition of \( \alpha \).
Table S5. Summary of the elastic properties (mainly the axial Young’s modulus) of spider silks from the literature. The data are organized by the spider species. Within the same species, the data are organized by the publication year. Note that the dragline silk is equivalent to the major ampullate silk. $E_\parallel$: axial Young’s modulus.

| Spider species | Silk type | Elastic properties | Measurement technique | Reference | Remarks (e.g., geometrical parameter, density, refractive index, technique limitations) |
|----------------|-----------|--------------------|-----------------------|-----------|----------------------------------------------------------------------------------|
| Nephila pilipes | Dragline | $E_\parallel = 0.003$-$0.035$ GPa | Stress-strain | Du2006 (ref 14) | The Young’s modulus are exceptionally low. |
| Nephila pilipes | Dragline | only stress-strain curve, no $E$ data | Stress-strain | Du2011 (ref 30) | Upon stretching, first strain-hardening, then strain-softening. |
| Nephila pilipes | Major ampullate | $E_\parallel = 2$-$16$ GPa | Stress-strain | Blamires2015 (ref 33) | Large variation. See Figure 3 of ref 33. |
| Nephila pilipes | Dragline | Ultimate tensile strength UTS = $1.030 \pm 0.176$ GPa Axial Young’s modulus $E_\parallel = 4$ GPa | Stress-strain | Kerr2018 (ref 36) | Naturally spun silk $E_\parallel$ is estimated from Figure 2 of ref 36 (left panel). |
| Nephila edulis | Major ampullate | $E_\parallel = 3.6$-$9.4$ GPa | Stress-strain | Madsen1999 (ref 20) | Depending on the starvation period and the reeling speed. Significant inter- and intraspecific differences. |
| Nephila edulis | Dragline | $E_\parallel = 10.4 \pm 0.8$ GPa | Stress-strain | Shao1999 (ref 21) | Spider silk in air; $D = 2.7$ μm; The modulus depends on the environment (e.g., air, water, solution, etc.) and humidity. |
| Nephila edulis | Major ampullate | $E_\parallel = 0.95$-$1.27$ GPa | Stress-strain | Riekel2000 (ref 23) | $D = 3.35 \pm 0.63$ μm; $E_\parallel$ increases with the drawing speed of silking. |
| Nephila edulis | Major ampullate | $E_\parallel = 8.71 \pm 1.50$ GPa | Stress-strain | Madsen2000 (ref 22) | $D = 3.57 \pm 0.53$ μm for silk diameter; $0.39 \pm 0.07$ for breaking strain, $1.29 \pm 0.16$ GPa for breaking stress, $8.71 \pm 1.50$ GPa for initial modulus, $217 \pm 45$ kJ kg$^{-1}$ for breaking energy, and $0.16 \pm 0.04$ GPa for yield stress. |
| Nephila edulis | Major ampullate | Axial longitudinal modulus $M_\parallel = 31.0 \pm 0.1$ GPa Transverse longitudinal modulus $M_\perp = 12.3 \pm 0.2$ GPa | BLS | Schneider2016 (ref 44) | Density, $\rho = 1250$ kg m$^{-3}$; $D = 2.0$ μm; $M_\parallel$ increases with strain, but $M_\perp$ is insensitive to strain. |
| Nephila clavipes | Dragline | $E_\parallel = 12.7$ GPa (controlled silking) $E_\parallel = 10.9$ GPa (natural silk) | Stress-strain | Zemlin1968 (ref 63) | |
| Nephila clavipes | Dragline | Initial, axial Young’s modulus $E_\parallel = 19.5$-$60$ GPa Average: $22$ GPa | Stress-strain | Cunniff1994 (ref 19) | $D = 3.0 \pm 0.62$ μm, depending on the drawing speed of silking. $E_\parallel$ depends on the drawing speed of silking. See Table 1 of ref 19 for additional mechanical properties of spider silks from previous studies. |
| Nephila clavipes | Dragline | $E_\parallel = 12.71$ GPa Lateral Young’s modulus $E_\perp = 0.579$ GPa Torsional modulus $G_\perp = 2.38$ GPa | Axial stress-strain test, lateral compression test, torsional test | Ko2004 (ref 25) | $D = 3.4$ μm |
| Nephila clavipes | Dragline | $E_t = 1-10$ GPa | Stress-strain | Elices2005 (ref 26) | $\rho = 1300$ kg m$^{-3}$; variation due to temperature, moisture, and time effects. |
|------------------|----------|------------------|--------------|-------------------|---------------------------------------------------------------------|
| Nephila plumipes | Dragline | Ultimate tensile strength UTS = 1.030 ± 0.206 GPa | Stress-strain | Kerr2018 (ref 36) | Naturally spun silk |
| Nephila clavata  | Dragline | $E_t = 10-13$ GPa | Stress-strain | Osaki2002 (ref 24) | Depending on the spider’s weight. |
| Nephila curvata  | Dragline | $E_t = 3.6$ GPa (natural silk) | Stress-strain | Zemlin1968 (ref 63) | |
| Pholcus phalangioides | Major ampullate | $E_t = 10.5 \pm 2.8$ GPa (FDL) $E_t = 11.2 \pm 1.0$ GPa (DDL) $E_t = 6.4 \pm 0.9$ GPa (WDL) | Stress-strain | Boutry2011 (ref 29) | FDL: forcibly silking (1 cm s$^{-1}$) DDL: spider dropping silking WDL: spider walking silking |
| Hololena adnexa  | Major ampullate | $E_t = 10.4 \pm 1.7$ GPa (FDL) $E_t = 18.9 \pm 3.3$ GPa (DDL) $E_t = 20.2 \pm 2.1$ GPa (WDL) | Stress-strain | |
| Peucetia viridans | Major ampullate | $E_t = 32.1 \pm 1.1$ GPa (FDL) $E_t = 22.8 \pm 2.8$ GPa (DDL) $E_t = 18.9 \pm 2.2$ GPa (WDL) | Stress-strain | |
| Achaearanea tepidariorum | Major ampullate | $E_t = 12.1 \pm 0.3$ GPa (DDL) $E_t = 9.3 \pm 1.0$ GPa (WDL) | Stress-strain | |
| Latrodectus hesperus | Major ampullate | $E_t = 18.4 \pm 0.9$ GPa (FDL) $E_t = 23.0 \pm 1.2$ GPa (DDL) $E_t = 17.3 \pm 1.2$ GPa (WDL) | Stress-strain | |
| Larinioides cornutus | Major ampullate | $E_t = 14.8 \pm 1.0$ GPa (FDL) $E_t = 17.3 \pm 1.6$ GPa (DDL) $E_t = 13.5 \pm 0.8$ GPa (WDL) | Stress-strain | |
| Araneus diadematus | Dragline | $E_t = 2.8$ GPa (natural silk) | Stress-strain | Calvert1988 (ref 18) | |
| Araneus diadematus | Cocoon | $E_t = 0.6$ GPa (natural silk) | Stress-strain | Madsen1999 (ref 20) | Depending on the reeling speed. Significant inter- and intraspecific differences. |
| Araneus diadematus | Major ampullate | $E_t = 3.6-7.3$ GPa | Stress-strain | Shao1999 (ref 21) | Spider silk in air; $D = 2.2-3.0$ µm; The modulus depends on the environment (e.g., air, water, solution, etc.) and humidity. |
| Araneus diadematus | Dragline | $E_t = 6.5$ GPa | Stress-strain | Heidebrecht2015 (ref 6) | |
| Araneus diadematus | Major ampullate | $E_t = 8 \pm 2$ GPa | Stress-strain | Zemlin1968 (ref 63) | $D = 3-4$ µm |
| Argiope aurantia | Dragline | $E_t = 9.7$ GPa (controlled silking) $E_t = 9.9$ GPa (natural silk) | Stress-strain | Zemlin1968 (ref 63) | Note the $E_t$ of MA silk is not the highest among the five types of silks (unexpected). See Table 1 of ref 27 for additional mechanical properties of spider silks. |
| Argiope aurantia | Dragline | $E_t = 34.00$ GPa | Stress-strain | Ko2004 (ref 25) | |
| Argiope argentata | Dragline | $E_t = 4.0$ GPa (natural silk) | Stress-strain | Zemlin1968 (ref 63) | |
| Argiope argentata (Fabricius 1775) | Major ampullate | $E_t = 8.0 \pm 0.8$ GPa | Stress-strain | Blackledge2006 (ref 27) | |
| Argiope argentata (Fabricius 1775) | Tubuliform | $E_t = 11.6 \pm 2.1$ GPa | Stress-strain | |
| Species                        | Type                     | $E_i$ (GPa) | Test Method                      | Notes                                                                 |
|-------------------------------|--------------------------|-------------|----------------------------------|----------------------------------------------------------------------|
| Argiope argentata             | Aciniform                | $E_i = 10.4 \pm 1.4$ | Stress-strain                   |                                                                        |
| Argiope argentata             | Capture spiral           | $E_i = 0.001 \pm 0.0001$ | Stress-strain                   |                                                                        |
| Argiope argentata             | Minor ampullate          | $E_i = 10.6 \pm 1.2$ | Stress-strain                   |                                                                        |
| Argiope trifasciata           | Dragline                 | $E_i = 1-10$ | Stress-strain                   | Elices2005 (ref 26)                                                   |
| Euprosthenops sp.             | Dragline                 | $E_i = 15.0 \pm 1.4$ | Stress-strain                   | Shao1999 (ref 21)                                                     |
| Latrodectus mactans           | Dragline                 | $E_i = 10.4 \pm 1.2$ | Stress-strain                   | Shao1999 (ref 21)                                                     |
| Parawixla audax               | Dragline                 | $E_i = 3.1$ | Stress-strain (natural silk)     | Zemlin1968 (ref 63)                                                  |
| Tetranychus urticae           | Spider mite silk fibers  | $E_i = 24 \pm 3$ (adult)  | AFM three-point bending test    | Hudson2013 (ref S4)                                                  |
| Liphistius mayalanus          | Dragline                 | $E_i = 4.8 \pm 0.5$ | Stress-strain                   | Swanson2009 (ref 28)                                                |
| Liphistius murphorium         | Dragline                 | $E_i = 3.7 \pm 0.1$ | Stress-strain                   |                                                                        |
| Cyroioagopus pagonus          | Dragline                 | $E_i = 3.7 \pm 0.2$ | Stress-strain                   |                                                                        |
| Pterinochilus marinus         | Dragline                 | $E_i = 2.6 \pm 0.1$ | Stress-strain                   |                                                                        |
| Poecilotheria regalis         | Dragline                 | $E_i = 4.3 \pm 1.0$ | Stress-strain                   |                                                                        |
| Aphenoplema seemani           | Dragline                 | $E_i = 1.6 \pm 0.2$ | Stress-strain                   |                                                                        |
| Phormictopus cancerides       | Dragline                 | $E_i = 2.9 \pm 0.1$ | Stress-strain                   |                                                                        |
| Grammastola rosea             | Dragline                 | $E_i = 1.0 \pm 0.1$ | Stress-strain                   |                                                                        |
| Cyclosternum fasciatum        | Dragline                 | $E_i = 1.4 \pm 0.5$ | Stress-strain                   |                                                                        |
| Seven species of spiders      | Dragline                 | $E_i = 2.5-8.6$ | Stress-strain                   | Madurga2015 (ref 32)                                                 |
| Several spider species        | Dragline                 | $E_i = 3-10$ | Stress-strain                   | Yarger2018 (ref 35)                                                  | Review paper |

$D$ varies due to temperature, moisture, and time effects. Spider silk in air; $D = 2.0 \mu m$; The modulus depends on the environment (e.g., air, water, solution, etc.) and humidity. Spider silk in air; $D = 2.1-3.2 \mu m$; The modulus depends on the environment (e.g., air, water, solution, etc.) and humidity. Depending on the type of spider silk and the sample. The modulus increases with crystallinity (good correlation). Seven species of spiders: Argiope aurantia, Aphonopelma seemani, Pisaura mirabilis, Caerostris darwini, Deinopis spinosa, Dolomedes tenebrosus, Kukulcania hibernalis. $E_i$ increases in the order.
Supplementary References

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