

Supplementary Information

for

“Flexible polymer-based liquid crystal sensor for label-free electro-optical recording of electrophysiological activity”

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Supplementary Note S1. Quantitative Analysis of Dielectric Interfaces

To justify the use of Eqn. 1 as a valid analytical approximation for the flexible optrode architecture, the parasitic reflections at each dielectric interface were quantified at the operating wavelength of 1550 nm. The complex refractive indices of the materials used at 1550 nm are summarized in Supplementary Table S1.

Calculation for Gold Mirror Reflectance:

The reflectance R_m at the interface between the PI alignment layer and the Gold mirror is calculated using the complex Fresnel equation for normal incidence:

$$R_m = \left| \frac{\tilde{n}_{Au} - n_{LC}}{\tilde{n}_{Au} + n_{LC}} \right|^2 = \left| \frac{(0.402 + 10.844i) - (1.70 + 0i)}{(0.402 + 10.844i) + (1.70 + 0i)} \right|^2 \quad (S1)$$

$$R_m = \frac{(-1.29821)^2 + (10.844)^2}{(2.10179)^2 + (10.844)^2} = \frac{1.6853 + 117.5923}{4.4175 + 117.5923} = \frac{119.2777}{122.0098} \approx 97.8\% \quad (S2)$$

This value represents the primary signal source for the optrode's optical read-out. The power reflection coefficient (R_i) at internal dielectric interfaces is calculated using the simplified Fresnel equation for non-absorbing media [1].
PI/LC Interface:

$$R_{PI/LC} = \left| \frac{(1.70 + 0i) - (1.60 + 0i)}{(1.70 + 0i) + (1.60 + 0i)} \right|^2 \approx 0.092\% \quad (S3)$$

LC/PEDOT:PSS Interface:

$$R_{LC/PEDOT} = \left| \frac{(1.60 + 0i) - (1.58 + 0.01i)}{(1.60 + 0i) + (1.58 + 0.01i)} \right|^2 \approx 0.005\% \quad (S4)$$

The total reflectance R_{total} measured by the system can be expressed as the sum of the primary mirror signal and the parasitic reflections:

$$R_{total} = R_m \cdot R_{||} + \sum R_{parasitic} \quad (S5)$$

Substituting the calculated values:

$$R_{total} \approx 0.978 \cdot R_{||} + 0.00189 \quad (S6)$$

where $R_{\parallel}$ represents the field-dependent parallel reflectance of the active LC layer, as modeled in Eqn. 1 of the main manuscript. The combined contribution of all internal parasitic reflections ($\approx 0.19\%$) is nearly three orders of magnitude smaller than the mirror reflectance ($\approx 97.8\%$). Consequently, the error introduced by neglecting these interfaces in Eqn. 1 (in the main manuscript) is less than 0.2% of the total signal, justifying the use of the simplified analytical model for both rigid and flexible optrode architectures.

Supplementary Note S2. Sensitivity Analysis of Structural Parameters

To address the uncertainty regarding the exact value of C and its dependence on the deformation mode, a parametric sensitivity analysis was performed. We utilized a heterogeneous composite beam model to calculate the position of the MNA (y_0) and the total flexural rigidity (EI) of the 7-layer optrode stack. The sensitivity of the device’s structural integrity to variations in C (ranging from 0.1 to 100) is detailed in Supplementary Table S2.

The results demonstrate that y_0 remains invariant, with a shift of less than 1 nm across three orders of magnitude of C . Even at $C = 10,000$, which results in a modulus higher than typical non-polymerized LCs, the MNA remains securely within the 5 μm LC cavity. This confirms that the structural protection of the LC layer is a robust consequence of the symmetry and dominance of the high-modulus polyimide substrates.

Supplementary Tables

Supplementary Table S1: Complex refractive indices of materials used at 1550 nm.

Material	Refractive Index ($\tilde{n} = n + ik$)	Function	Ref
Polyimide	$\approx 1.70 + 0i$	Alignment Layer	[2]
PEDOT:PSS/EG	$\approx 1.58 + 0.01i$	Transparent Electrode	[3]
DHF LC*	$(1.58 - 1.69) + 0i$	Transduction Component	[4]
Gold (Au)	$0.402 + 10.844i$	Mirror	[5]

**Note: The $\tilde{n}$ of the DHF LC mixture is anisotropic, representing the uniaxial range between the ordinary index ($n_o \approx 1.58$) and the extraordinary index ($n_e \approx 1.69$) at 1550 nm. An effective isotropic average of $n_{LC} = 1.60$ was adopted for calculations to represent the unperturbed bulk state.*

Supplementary Table S2: Sensitivity of MNA position (y_0) and flexural rigidity (EI) to variations in the LC effective modulus.

Constant C	$E_{LC,eff}$ (Pa)	y_0 (μm)	Total EI ($N \cdot m^2$)	ΔEI (%)
0.1	40	12.868	2.09×10^{-8}	0.000
1.0	400	12.868	2.09×10^{-8}	0.000
10	4,000	12.868	2.09×10^{-8}	0.000
100	40,000	12.868	2.09×10^{-8}	0.001

Note: Calculations utilize a 25.265 μm total stack thickness. y_0 is measured from the bottom substrate. Baseline values are highlighted.

Supplementary Table S3: Mechanical properties of the material model used in simulations. All values were determined experimentally in this work, except for Poisson’s ratio, which was taken from [6].

Parameter	Symbol	Value	Unit
Young’s modulus	E	1.5	GPa
Poisson’s ratio	η	0.34	-
Initial yield stress	σ_{ys0}	45	MPa
Strength coefficient	k	950	MPa
Hardening exponent	n	0.4	-

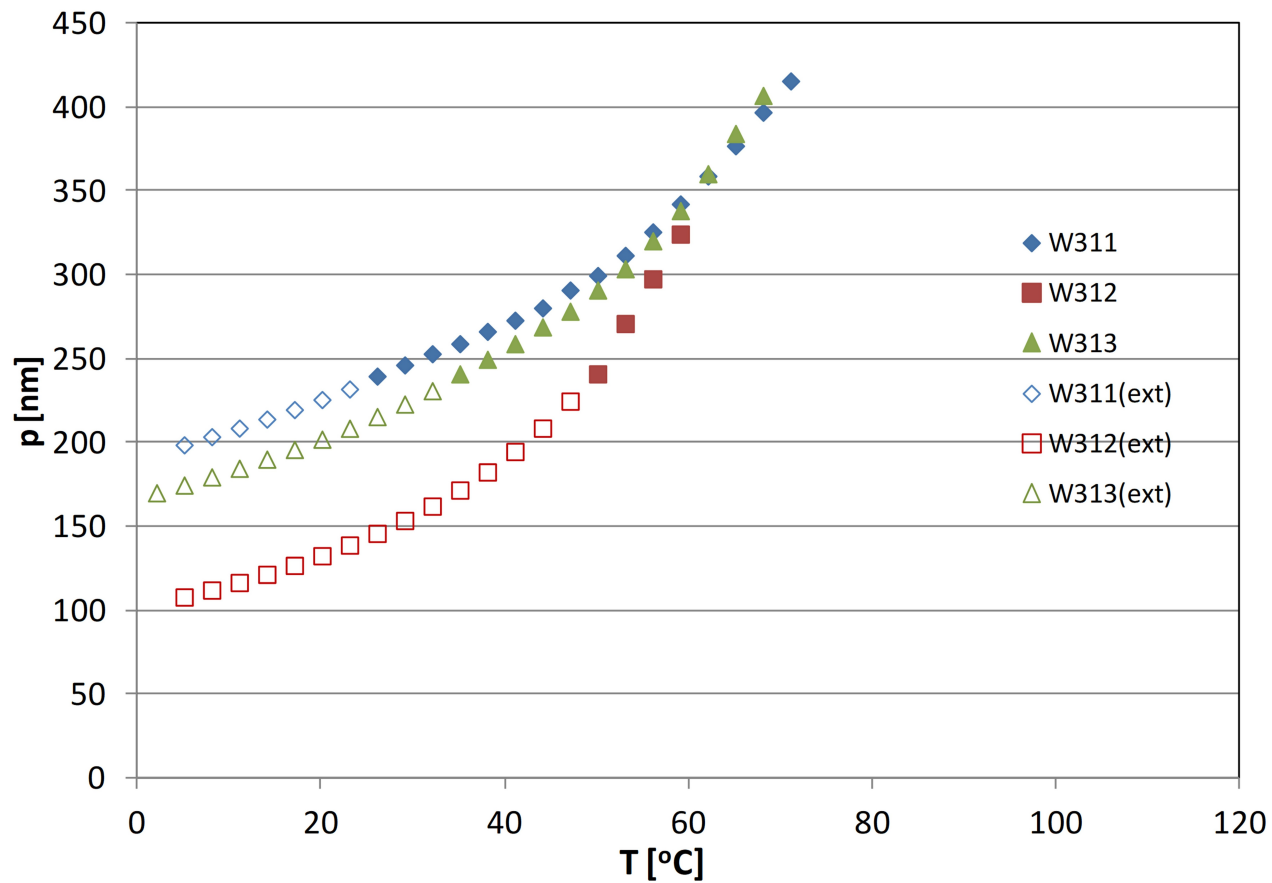
Supplementary Table S4: Material properties of the materials used in calculations.

Material	Thickness (μm)	Young’s Modulus (GPa)
PI substrates	10.0	1.50
LC layer	5.0	4×10^{-7}
PI alignment layers	0.060	1.50
PEDOT:PSS/EG	0.065	2.40
Au mirror	0.080	40

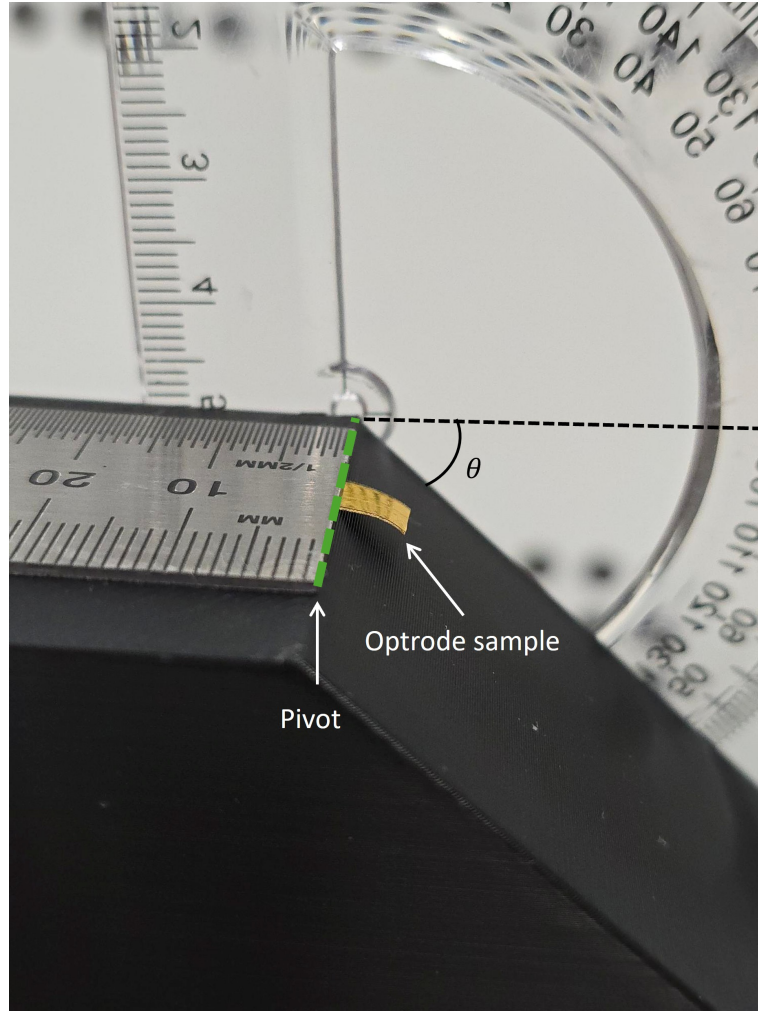
Supplementary Table S5: Quantitative noise and sensitivity parameters of the camera-based benchtop characterization system.

Parameter	Formula / Definition	Value
Calibration factor	C_{cal}	$5.1 \times 10^{-5} \text{ mV}^{-1}$
Raw baseline noise	σ_{raw}	1.182051×10^{-4}
Raw SNR	$\text{SNR}_{\text{raw}} = 10 \log_{10} \left(\frac{\sigma_{\text{signal}}^2}{\sigma_{\text{noise}}^2} \right)$	0.16 dB
Raw Noise-Equivalent Voltage	$\text{NEV}_{\text{raw}} = \frac{\sigma_{\text{raw}}}{C_{\text{cal}}}$	2.32 mV
Averaged baseline noise ($N = 7$)	$\sigma_{\text{averaged}} = \frac{\sigma_{\text{raw}}}{\sqrt{N}}$	4.468×10^{-5}
Averaged Noise-Equivalent Voltage	$\text{NEV}_{\text{averaged}} = \frac{\sigma_{\text{averaged}}}{C_{\text{cal}}}$	0.88 mV

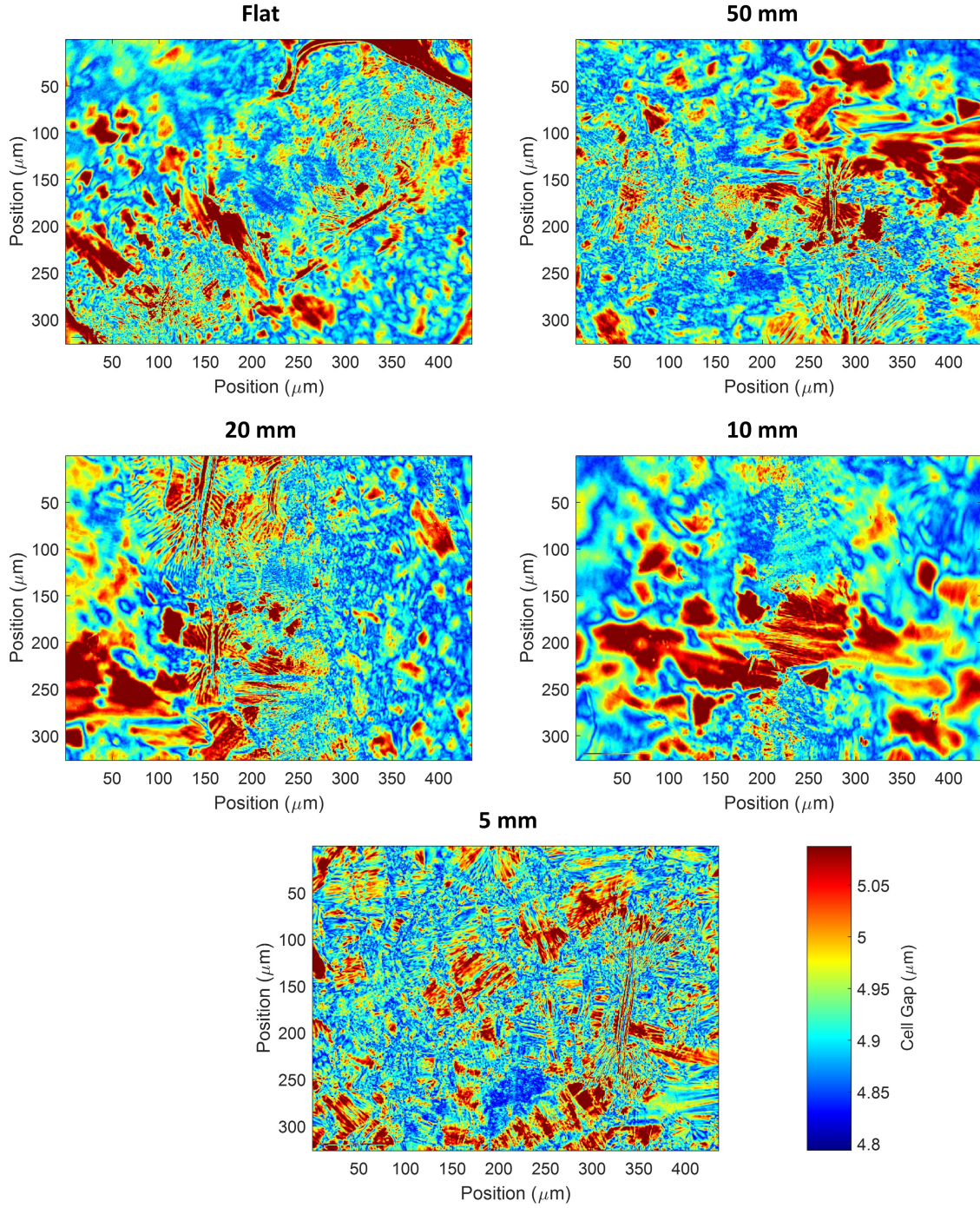
Supplementary Figures



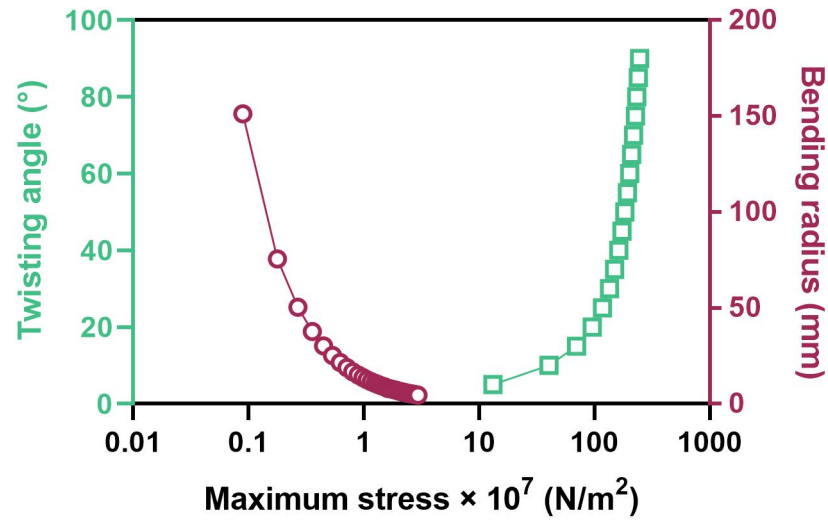
Supplementary Figure S1: Dependence of helical pitch on temperature for DHF LC mixtures W311, W312, W313, and W314. W311 is the one used in this study.



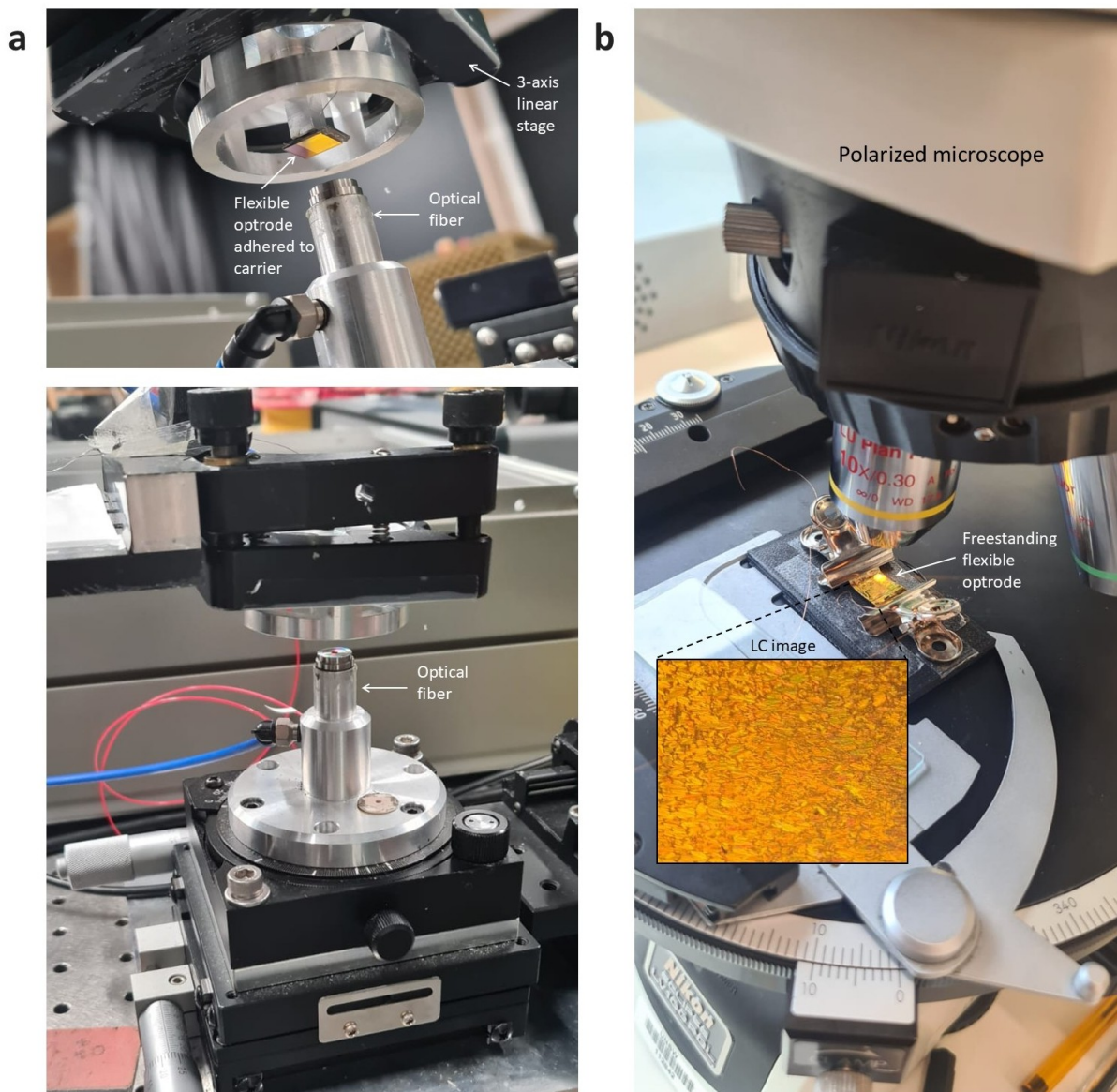
Supplementary Figure S2: A flexural testing apparatus built in accordance with ASTM D1388-18, utilizing a 3D-printed platform with a precision 41.5° incline marker. Three representative samples ($n = 3$) were tested and fabricated with the identical $25\ \mu\text{m}$ stack sequence described in the main manuscript. Each specimen was advanced along the horizontal platform until its leading edge reached the inclined plane, at which point the overhanging length (L) was recorded and the subsequent bending length (C) and flexural rigidity (G) were calculated.



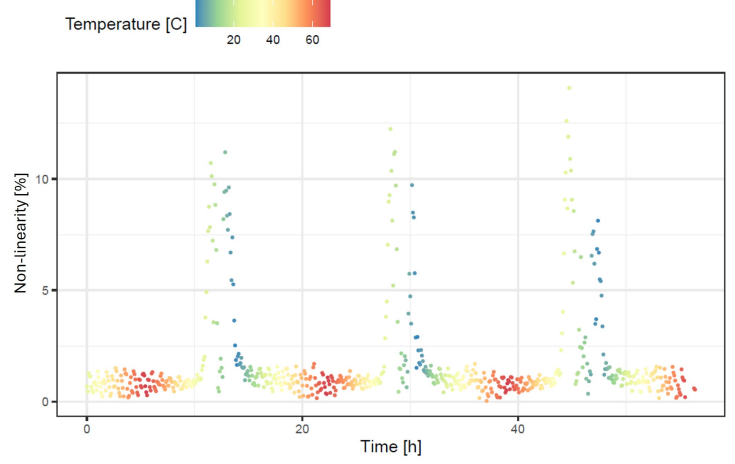
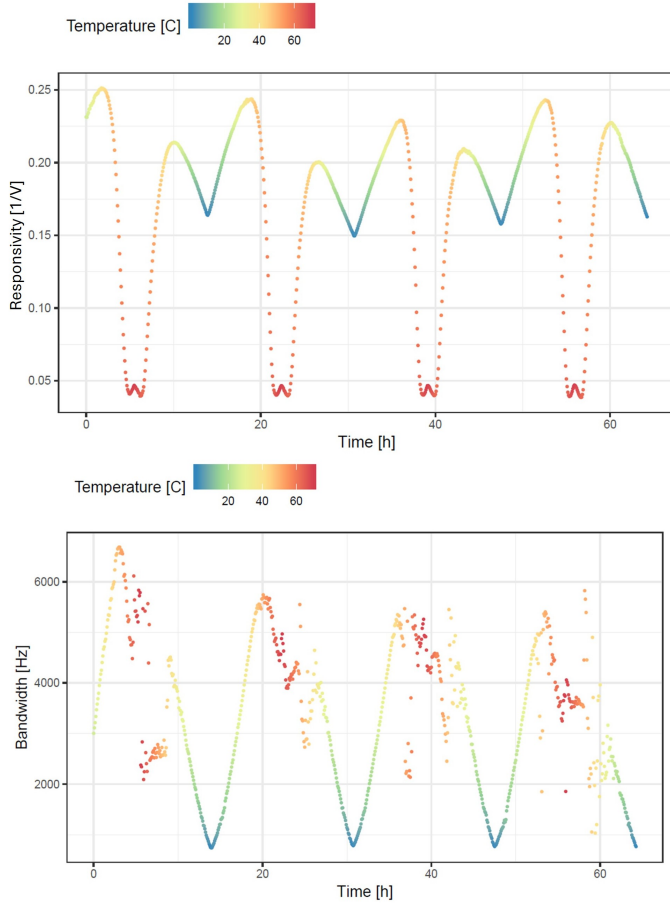
Supplementary Figure S3: Full-field 2D spatial heatmap of the cell gap d reconstructed phase-shifting algorithm across five bending radii ($r = 50, 20, 10, 5$ mm, and flat state). The map illustrates high spatial uniformity centered at $5.0 \pm 0.1 \mu\text{m}$, with texture patterns representing the natural domain structure of the Deformed Helix Ferroelectric (DHF) phase.



Supplementary Figure S4: FEA predicted maximum mechanical stress within a flexible optrode device with different bending radii and twisting angles.

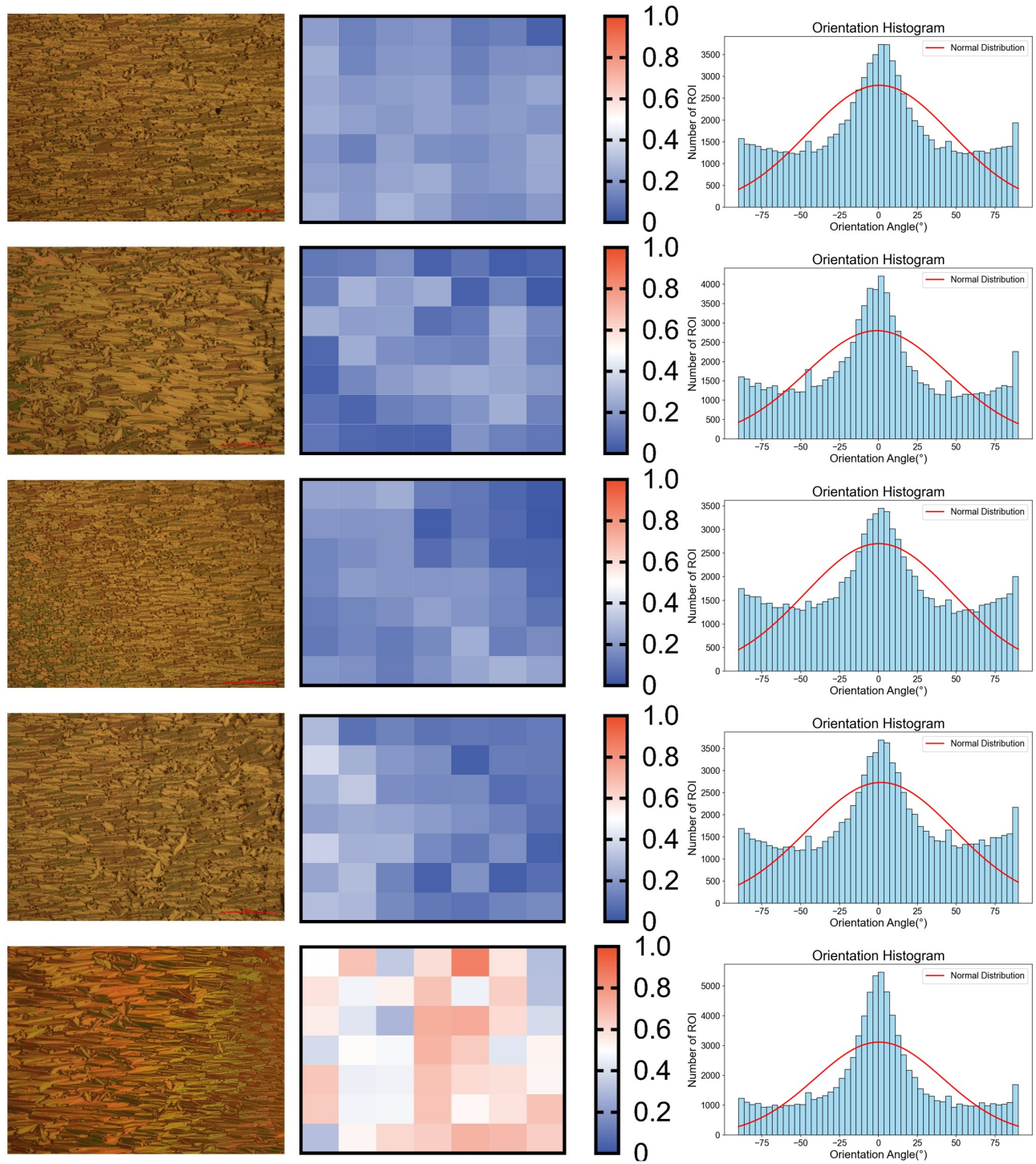


Supplementary Figure S5: Images of the optical setups used to characterize the optrode devices. **a** Free-space setup with an optic fiber and a photodetection system for unreleased flexible optrodes. **b** Polarized optical microscope with camera to image freestanding flexible optrodes.

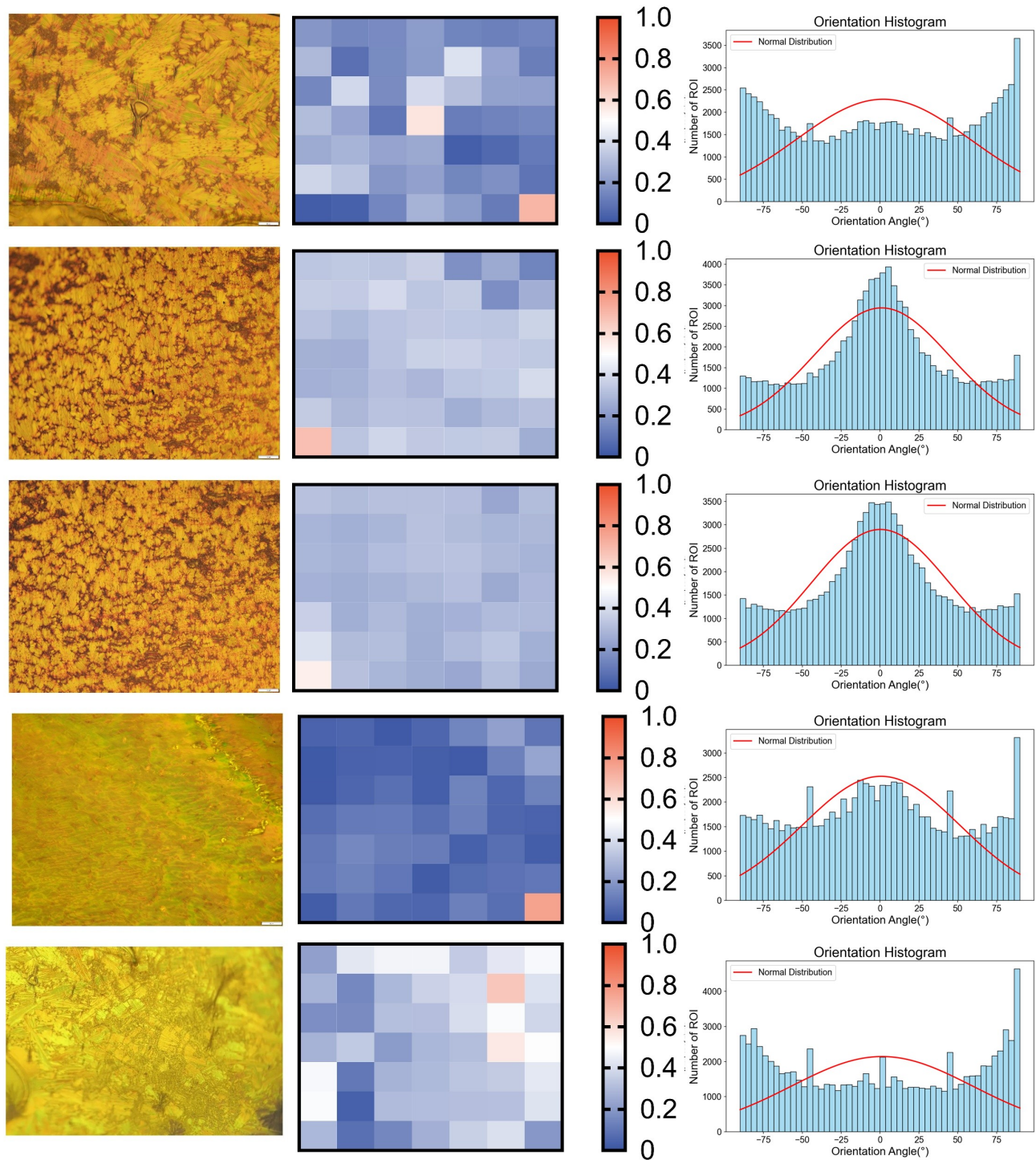


Device property	25°C	37°C
Responsivity [1/V]	0.21±0.01	0.22±0.02
Bandwidth [kHz]	2.91±0.89	4.33±0.44
Non-linearity [%]	1.14±0.41	0.91±0.12

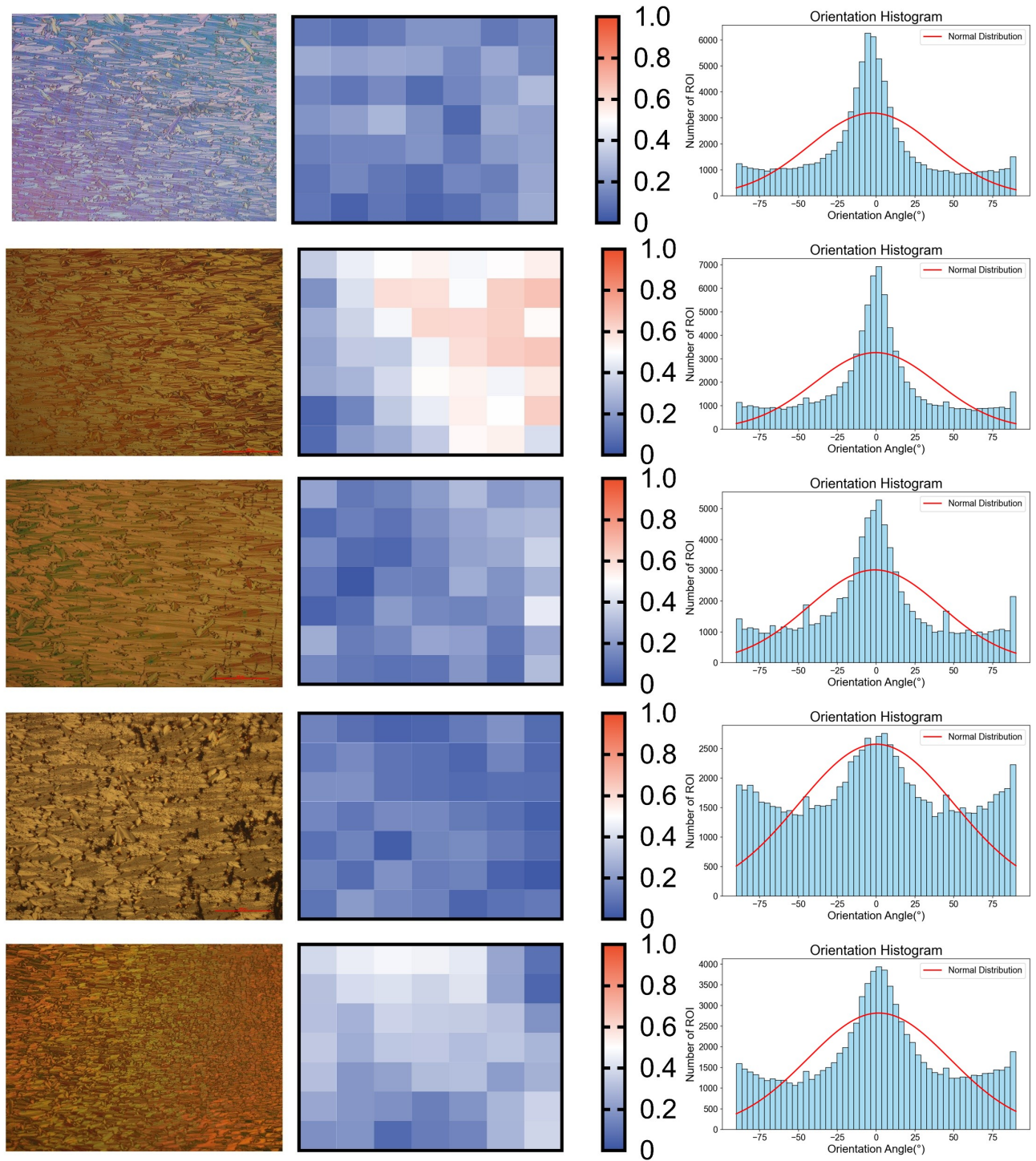
Supplementary Figure S6: Temperature-dependent relative responsivity (1/V), bandwidth (Hz), and linearity (%) of the DHF LC transducer over time. Data was measured at 5-minute intervals over a 60-hour duration. Bottom right: A summary table directly comparing key electro-optical properties at room temperature (25°C) and core body temperature (37°C), where the reported data represent mean $\pm$ standard deviation (s.d.) ($n = 8$ measurements at each temperature).



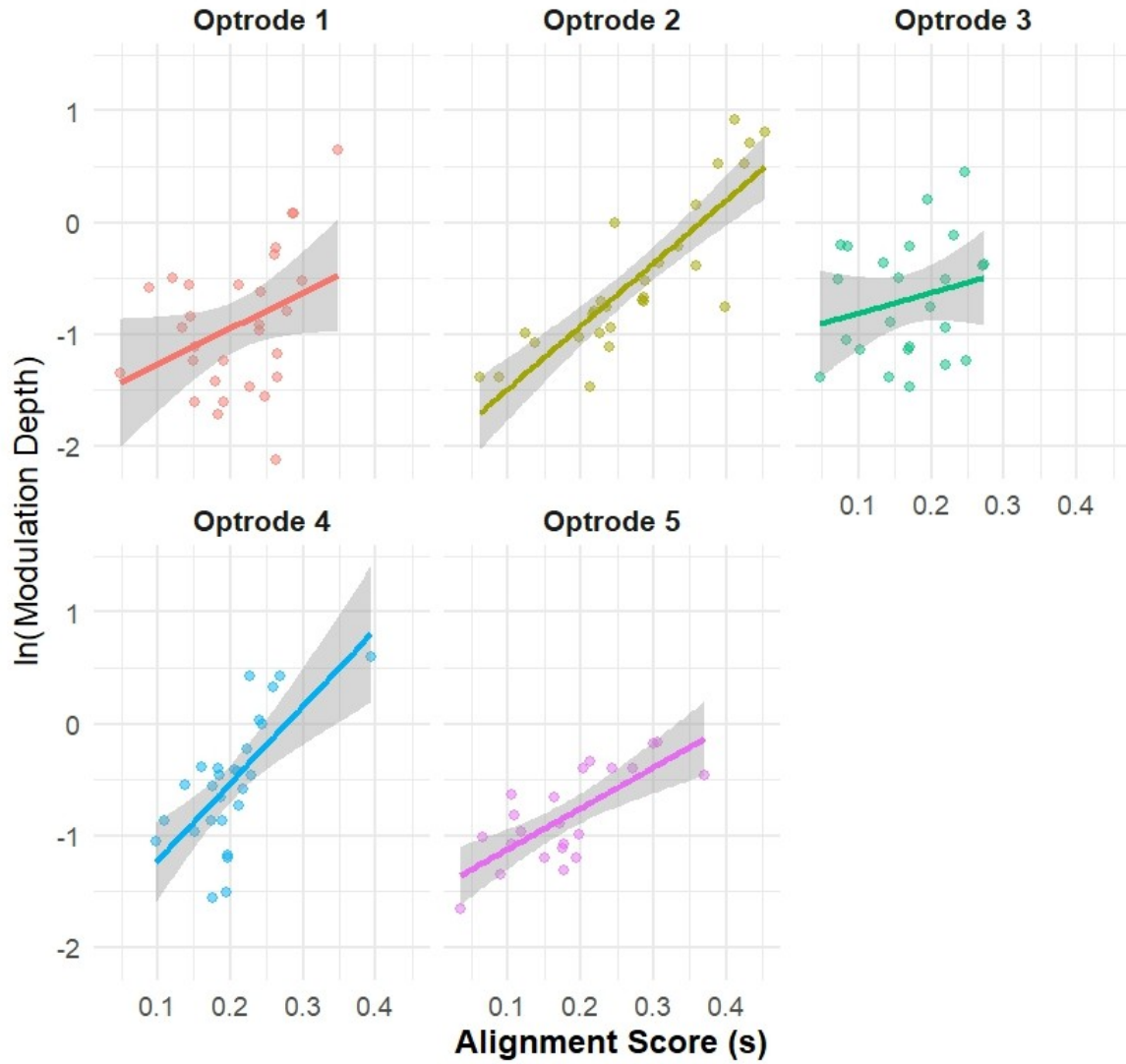
Supplementary Figure S7: Representative polarized optical micrographs of unreleased flexible optrodes with their corresponding alignment heatmaps and orientation angle histograms. The heatmaps depict local alignment scores across subdivided regions of interest, while the histograms summarize the orientation angle distributions.



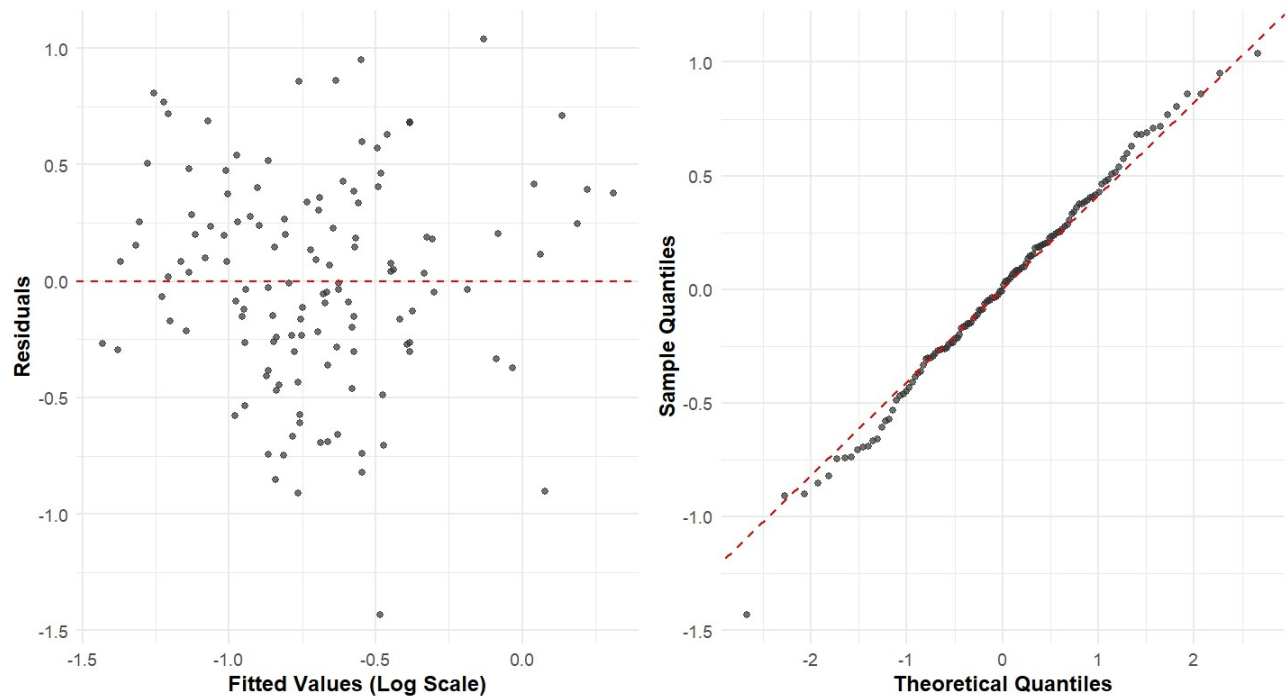
Supplementary Figure S8: Representative polarized optical micrographs of released flexible optrodes with their corresponding alignment heatmaps and orientation angle histograms. The heatmaps depict local alignment scores across subdivided regions of interest, while the histograms summarize the orientation angle distributions.



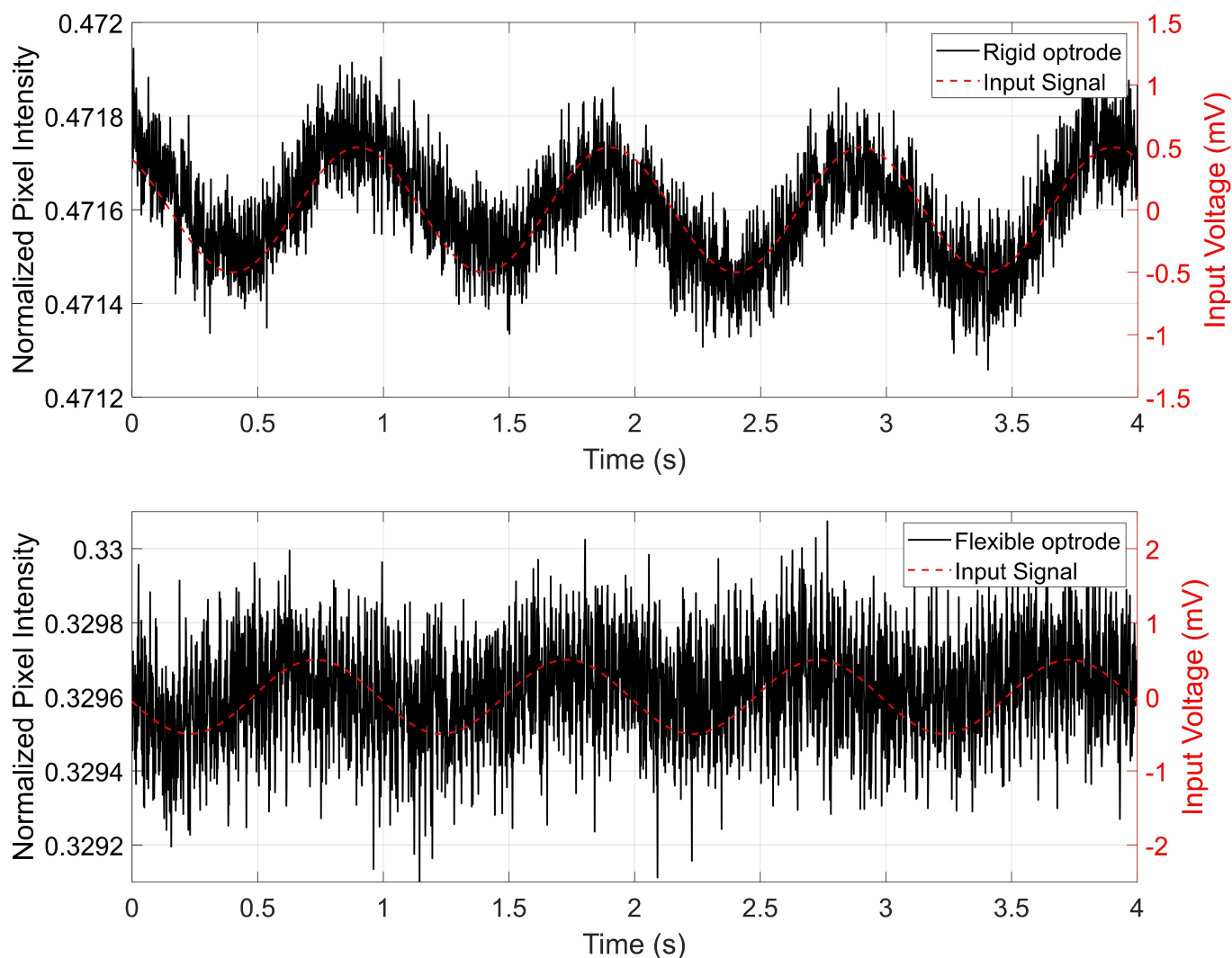
Supplementary Figure S9: Representative polarized optical micrographs of rigid optrodes with their corresponding alignment heatmaps and orientation angle histograms. The heatmaps depict local alignment scores across subdivided regions of interest, while the histograms summarize the orientation angle distributions.



Supplementary Figure S10: Relationship between LC alignment score and device modulation depth. Solid lines represent fits derived from the linear mixed-effects (LME) model applied to log-transformed data, with shaded regions indicating 95% confidence intervals for each device. The global fixed-effect slope was statistically significant ($\beta = 4.15$, $SE = 0.83$, $t(3.02) = 4.98$, $p = 0.015$).



Supplementary Figure S11: Diagnostic plots for the LME model. Left: Residuals plotted against fitted values (log-scale) demonstrate homoscedasticity, with residuals randomly distributed around the zero-line (dashed). Right: Normal Q-Q plot of the residuals showing good agreement with the theoretical normal distribution, indicating that the model assumptions of normality are satisfied.



Supplementary Figure S12: Detection of small signals using optrode devices via high-speed camera. A $1 \text{ mV}_{\text{p-p}}$, 1 Hz sinusoidal signal was applied to a rigid optrode (top) and a flexible optrode (bottom), and the output was measured as a function of pixel intensity (non-filtered data). The modulation depth for rigid and flexible optrodes is 0.026% and 0.016%, respectively.

Caption for Supplementary Movie S1. Polarized optical microscopy video showing the dynamic response of the deformed helix ferroelectric liquid crystals (DHF LCs) to an applied sinusoidal electric field ($2 V_{p-p}$, 1 Hz).

References

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