SUPPLEMENTARY INFORMATION to:

Multi-responsive poly-catecholamine nanomembranes

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- Structural and elastic properties of poly-catecholamine pristine membranes
- Moisture response of the polydopamine film PDA 200c transferred onto a silicon substrate
- Photoactuation dynamics of poly-catecholamine membranes irradiated with the red laser
- Photoactuation dynamics of polydopamine membranes irradiated with the white light
- Brillouin Light Scattering results
- Cyclic voltammograms of polycatecholamine films

Structural and elastic properties of poly-catecholamine pristine membranes

Table S1. Structural and elastic properties of poly-catecholamine pristine membranes determined at 23°C and RH = 30%. (ρ – mass density, d – thickness, E – Young modulus, σ_0 – residual stress, D – rigidity modulus). Values marked with * are taken from Ref.[60,61].

Sample	ρ (kg/m^3)	d (nm)	E (GPa)	σ_0 (MPa)	D ($\times 10^{-15}$ N·m)
PDA 15c	1866 ± 32	21.2 ± 0.5	8.1 ± 0.4	12.6 ± 1.0	7.1 ± 0.5
PDA 200c	1950 ± 60	18.9 ± 0.3	11.0 ± 0.5	19.5 ± 1.9	6.8 ± 0.3
PdDOPA	780 ± 40*	28.0 ± 1.0*	3.5 ± 0.4	3.3 ± 2.7	7.0 ± 0.8
PNE	1380 ± 70*	17.0 ± 1.0*	16.3 ± 2.1	0.5 ± 2.2	7.3 ± 1.3

Moisture response of the polydopamine film PDA 200c transferred onto a silicon substrate

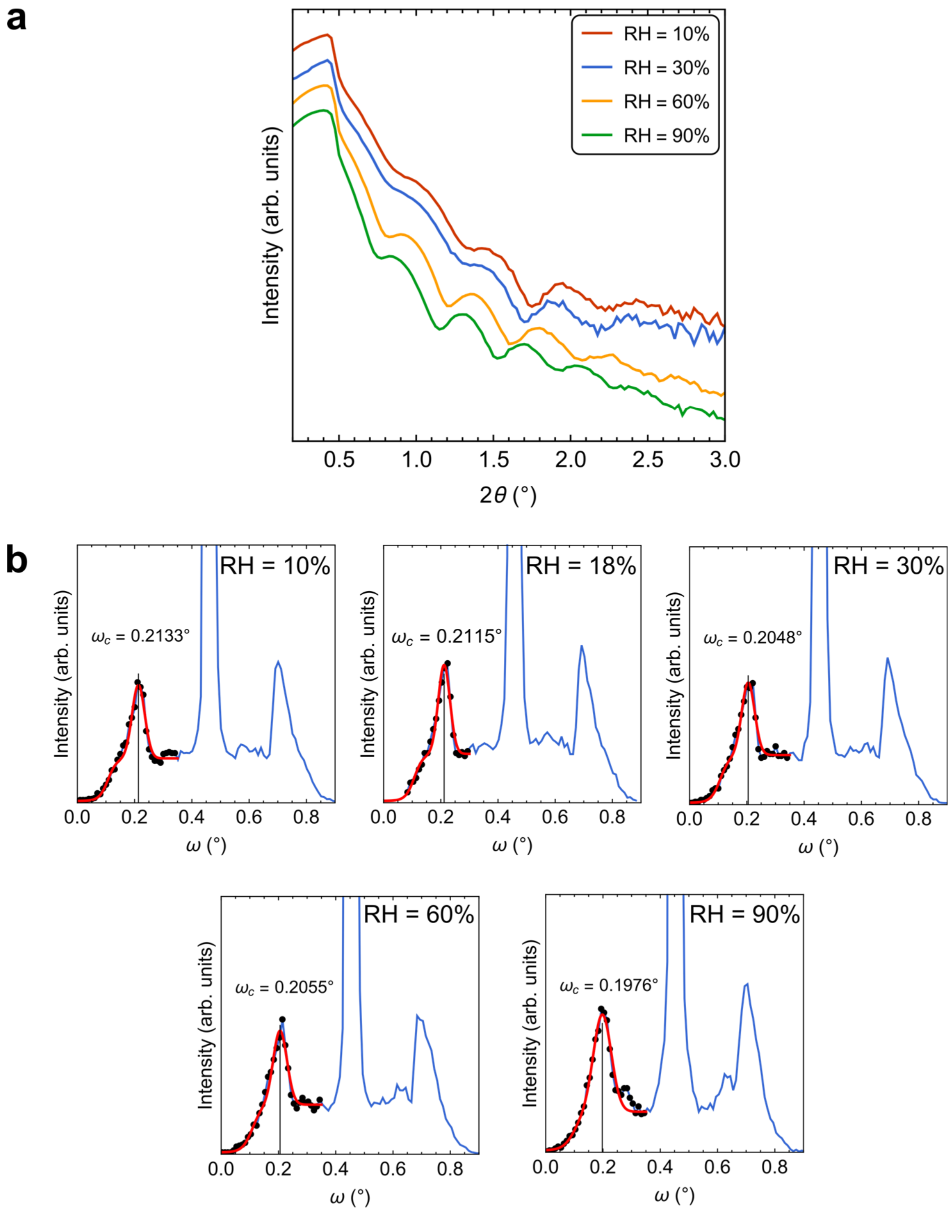

Figure S1. (a) Comparison of X-ray reflectometry spectra measured at different relative humidities (RH) for PDA 200c transferred onto Si substrate. The change in the oscillation period clearly shows the systematic variation in the film thickness. The spectra are shifted with respect to each other on the intensity axis for better comparison. (b) Rocking curves taken at $2\theta = 0.9°$ and different RH for PDA 200c transferred onto Si substrate. The apparent Yoneda wings around the specular reflection at $\omega = 0.45°$ allowed for the determination of the critical angle ω_c and, thus, the film density.

Photoactuation dynamics of poly-catecholamine membranes irradiated with the red laser

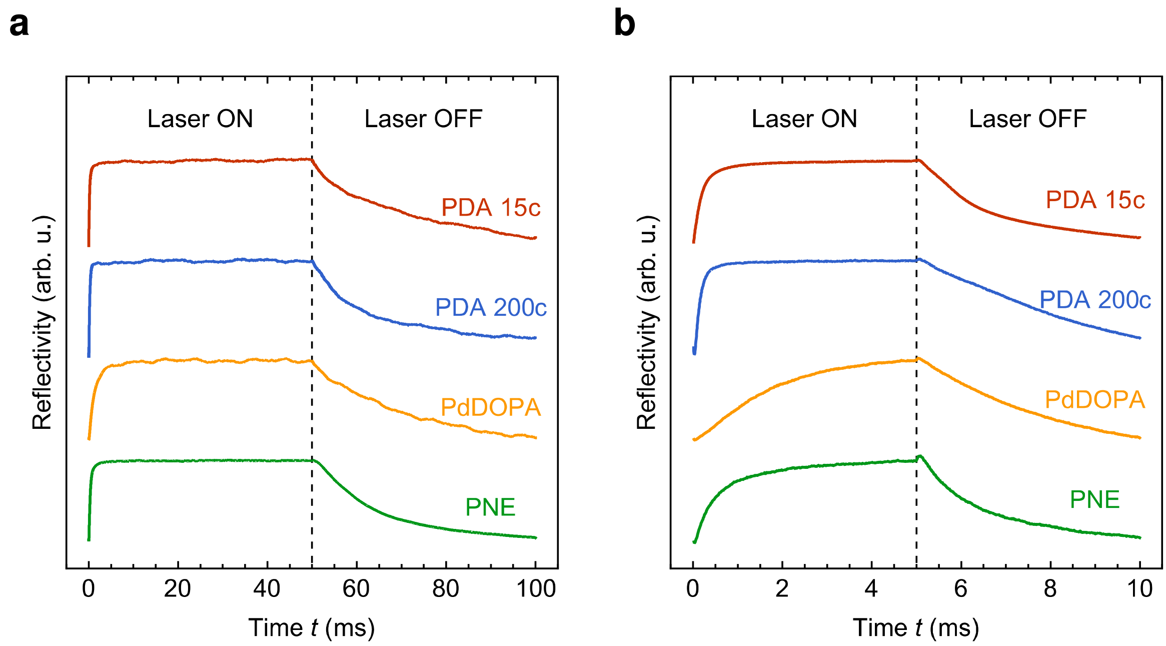

Figure S2. Photoactuation dynamics of poly-catecholamine membranes irradiated with the red laser (660 nm) at a repetition rate of (a) 10 Hz, and (b) 100 Hz.

Photoactuation dynamics of polydopamine membranes irradiated with white light

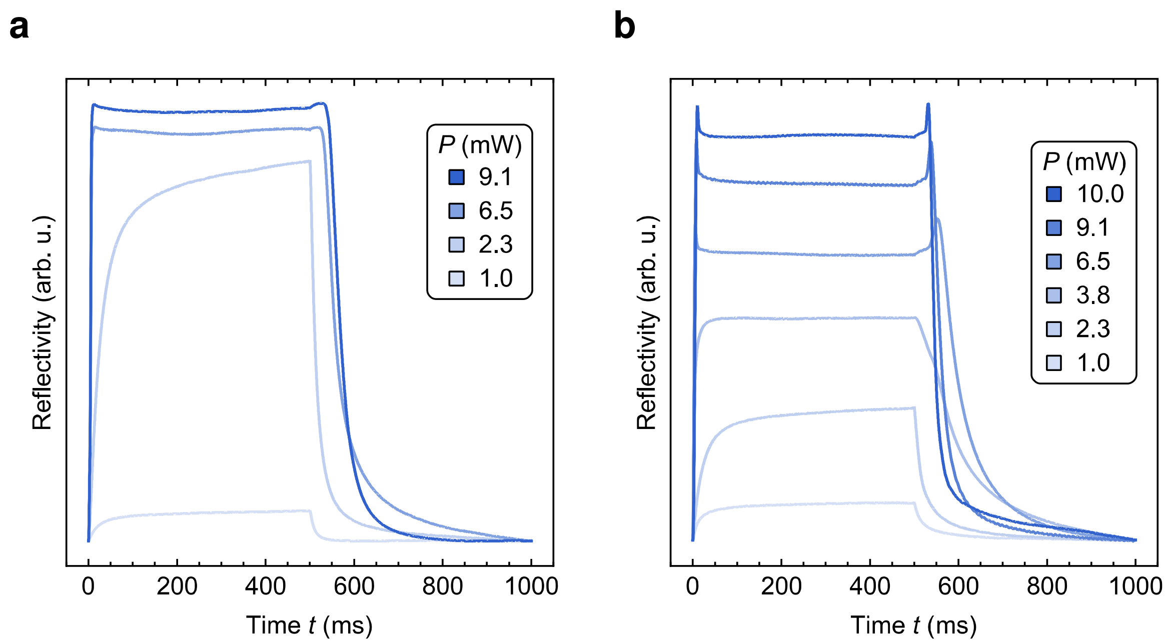

Figure S3. Photoactuation dynamics of PDA membranes irradiated with the white light at the repetition rate of 1 Hz for various power levels P. Results shown in (a) and (b) correspond to different PDA 200c membranes of the same sample.

Brillouin Light Scattering results

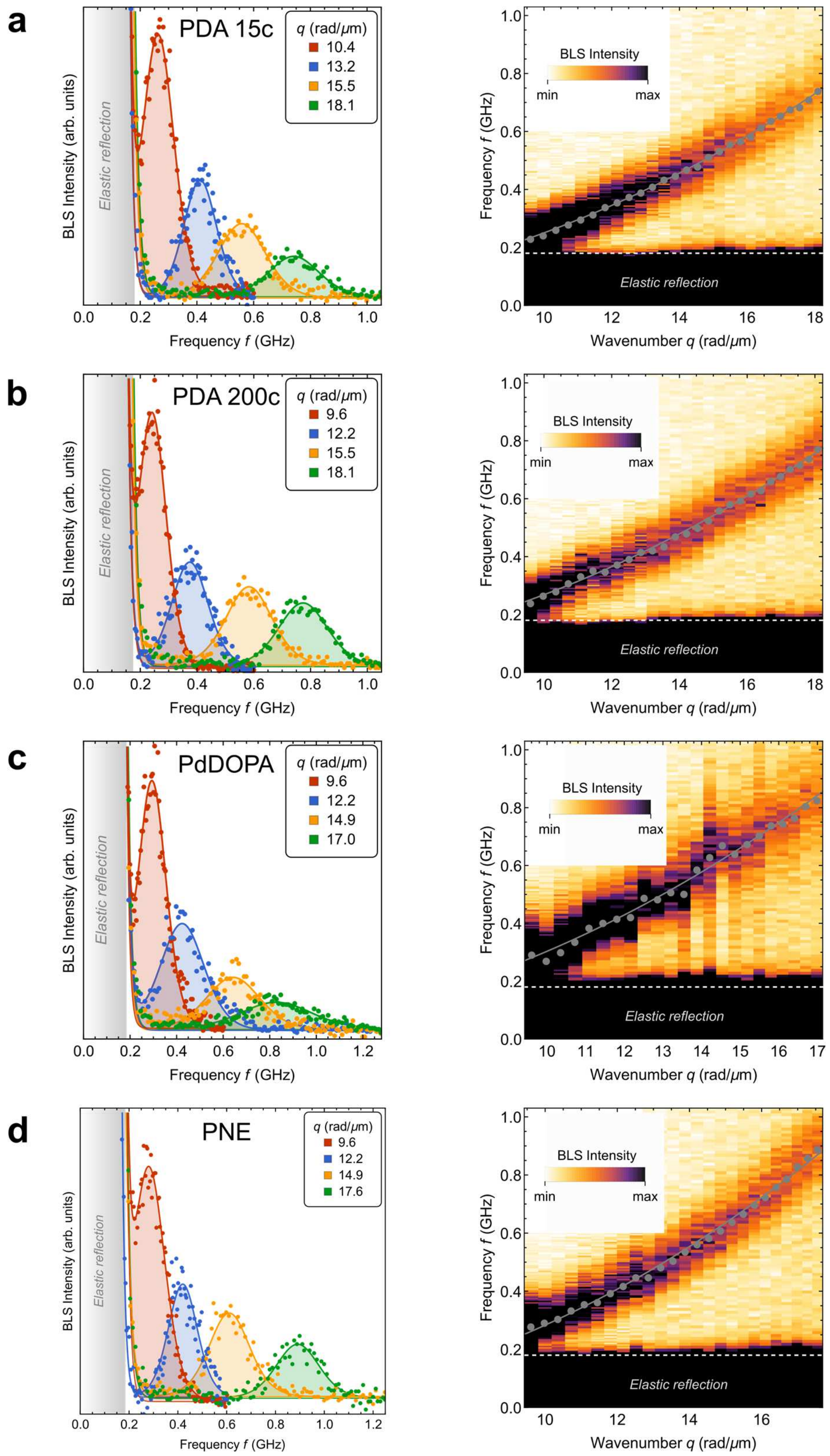

Figure S4. Micro-Brillouin Light Scattering results of (a) PDA 15c, (b) PDA 200c, (c) PdDOPA, and (d) PNE. The left panels show exemplary BLS spectra of antisymmetric, flexural Lamb waves (A0 mode). The right panels show color-coded dispersion relations of poly-catecholamine membranes. The grey points display the determined maxima of the scattering peak, and the continuous grey lines show fitting according to Eq. 1 in the main text.

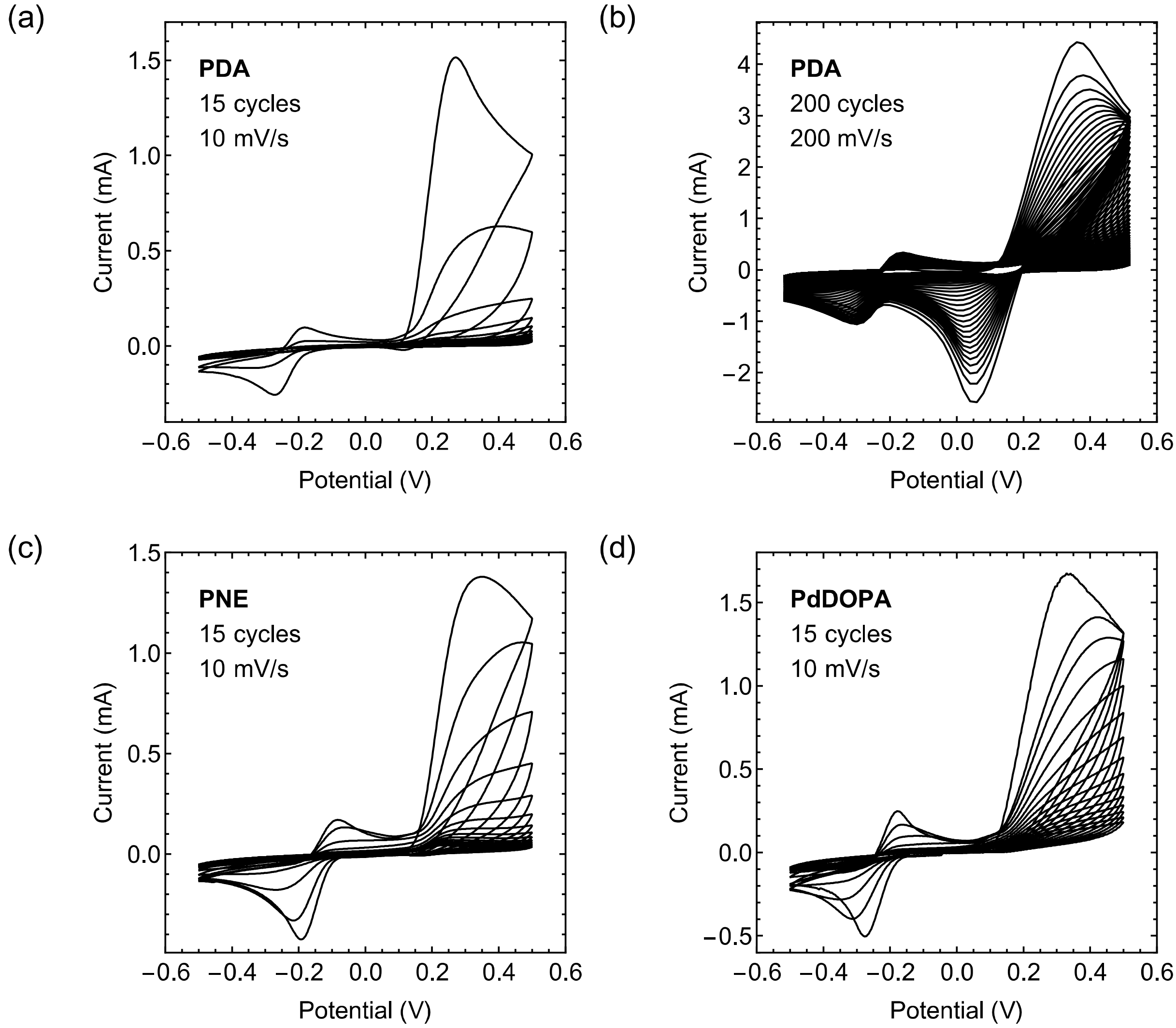

Figure S5. Cyclic voltammograms of polycatecholamine films in phosphate buffer pH 7. The potential was swept between +0.5 and –0.5 V, starting and ending at 0 V. (a) PDA film formation with 15 cycles and 10 mV/s scan rate, (b) PDA formation with 200 cycles and 200 mV/s scan rate, (c) PNE film formation with 15 cycles and 10 mV/s scan rate (d) PdDOPA formation with 15 cycles and 10 mV/s scan rate. The first oxidation peak for the reaction from catechol to quinone form of dopamine molecule can be distinguished at 0.3 V. The respective reduction peak is only visible for the 200 cycles PDA film due to the higher scan rate, resulting in higher currents. In the case of the PDA film synthesized with 200 cycles and 200 mV/s, higher currents can be observed due to the higher scan rate. The higher scan rate results in less time for the molecule to undergo the intrinsic change, which leads to PDA formation. Therefore, 200 cycles were performed at the higher scan rate to yield a PDA film with nearly the same thickness as the 15 cycle confirmed with X-ray reflectivity and atomic force microscopy measurements.