Electronic Supplementry Information: Transcription dynamics stabilizes nucleus-like layer structure in chromatin brush

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S1   Force balance equation and Maxwell construction

We here derive the force balance equation of a DNA brush and show that the Maxwell construction provides the necessary condition of the coexistence of swollen and collapsed DNA chains. We treat DNA chains that are end-grafted to a surface with grafting density σ. These DNA chains are composed of N chain segments of Kuhn length l_{eff}. The free energy per unit area of a DNA brush has the form

$$
\begin{aligned}
\frac{f_{\mathrm{ela}}}{T} &= \frac{3}{2}\frac{\sigma h^2}{N l_{\mathrm{eff}}^2} + \left[\frac{1}{2}w_{\mathrm{on}}\Phi_{\mathrm{on}}^2 + w_{\mathrm{int}}\Phi_{\mathrm{on}}\Phi_{\mathrm{off}} + \frac{1}{2}w_{\mathrm{off}}\Phi_{\mathrm{off}}^2 + \frac{1}{3}u\Phi_{\mathrm{on}}^3\right]h \\
&\quad + \frac{\Pi_{\mathrm{app}}}{T}h + \frac{1}{2}K(\nabla h)^2 + \frac{1}{2}\chi_\perp \sigma^{-1}h(\nabla\Phi_{\mathrm{on}})^2.
\end{aligned}
\tag{S1}
$$

The first term of eq. (S1) is the free energy due to entropic elasticity of DNA chains per unit area. The second term is the free energy due to the interactions between DNA chain segments. The third term is the work done by applied pressure. The fourth term is the elastic free energy of the grafting surface. The fifth term is the gradient energy, which is the excess interaction energy due to the gradient of the brush height and the nucleosome occupancy (see also ref.[1]). This free energy is minimized with respect to the brush height h and the nucleosome occupancy n_{his} is determined by the transcription dynamics and the dynamics of nucleosome assembly.

The Kuhn length l_{eff} has the form

$$
l_{\mathrm{eff}} = l_{\mathrm{a}}(1 - \gamma n_{\mathrm{his}}),
\tag{S2}
$$

where l_{a} is the Kuhn length of vacant DNA chain segments and γ is the constant that accounts for the fact that DNA chain segments are reeled around histone proteins when nucleosomes are assembled. The second virial coefficients, w_{on}, w_{int}, and w_{off}, account for the (nucleosome)-(nucleosome) interactions, the (nucleosome)-(vacant DNA segment) interactions, and the (vacant DNA segment)-(vacant DNA segment) interactions, respectively.

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The (nucleosome)-(nucleosome) interactions are attractive ($w_{on} < 0$), and the (vacant DNA segment)-(vacant DNA segment) interactions are repulsive ($w_{off} > 0$). u is the third virial coefficient that accounts for the three-body interactions between nucleosomes. Φ_{on} and Φ_{off} are the local concentrations of nucleosomes and vacant DNA chain segments and have the forms

$$\Phi_{on} = \frac{\sigma N}{h} n_{his} \tag{S3}$$

$$\Phi_{off} = \frac{\sigma N}{h}(1 - n_{his}). \tag{S4}$$

K is the bending rigidity of the grafting surface and ∇ is the two dimensional gradient in the lateral direction. $\chi_{\perp}$ is the constant that accounts for the gradient energy and is $-w \ (= 2w_{int} - w_{on} - w_{off})$ in a lattice model.[1]

The fourth and fifth terms of eq. (S1) are interfacial terms that are zero in the bulk of domains. Minimizing the free energy, eq. (S1), with respect to the brush height h leads to the form

$$\frac{\Pi_{app}}{T} = -\frac{3\sigma h}{N l_{eff}^2} + \frac{1}{2}w_{on}\Phi_{on}^2 + w_{int}\Phi_{on}\Phi_{off} + \frac{1}{2}w_{off}\Phi_{off}^2 + \frac{2}{3}u\Phi_{on}^3. \tag{S5}$$

Eq. (S5) is equal to eq. (10) in the main article. The fifth term is significant only for relatively large values of the nucleosome occupancy, $n_{his} \sim 1$. Rewriting the second, third, and fourth terms leads to the form

$$\frac{\Pi_{app}}{T} = -\frac{3\sigma h}{N l_{eff}^2} + \frac{1}{2}w\frac{N^2\sigma^2}{h^2}(n_{his} - n_+)(n_{his} - n_-) + \frac{2}{3}u\frac{N^3\sigma^3}{h^3}, \tag{S6}$$

where w, n_+, and n_- have the forms

$$w = w_{on} + w_{off} - 2w_{int} \tag{S7}$$

$$n_{\pm} = \frac{w_{off} - w_{int} \pm \sqrt{w_{int}^2 - w_{on}w_{off}}}{w}. \tag{S8}$$

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We used eq. (S6) to derive fig. 3 in the main article.

The brush height h and the nucleosome occupancy n_{his} is a function of position at interfaces between domains and the fourth and fifth terms of eq. (S1) are not zero at interfaces. Applying the variational principle with respect to the brush height h to the free energy density leads to the force balance equation

$$-K\nabla^2 h - \chi_\perp \sigma^{-1} h \Phi_{\mathrm{on}}'^2(h)\nabla^2 h - \frac{1}{2}\chi_\perp \sigma^{-1}\frac{d}{dh}(h\Phi_{\mathrm{on}}'^2(h))(\nabla h)^2 - \frac{\Pi_\perp(h)}{T} = 0. \qquad (S9)$$

The bulk component of osmotic pressure $\Pi_\perp(h)$ has the form

$$\frac{\Pi_\perp(h)}{T} = -\frac{3\sigma h}{N l_{\mathrm{eff}}^2} + \frac{1}{2}w\frac{N^2\sigma^2}{h^2}(n_{\mathrm{his}} - n_+)(n_{\mathrm{his}} - n_-) + \frac{2}{3}u\frac{N^3\sigma^3}{h^3} - \frac{\Pi_{\mathrm{app}}}{T}. \qquad (S10)$$

We used a relationship

$$
\begin{aligned}
\delta\left[\frac{1}{2}\chi_\perp\sigma^{-1}h(\nabla\Phi_{\mathrm{on}})^2\right] &= \frac{1}{2}\chi_\perp\sigma^{-1}\delta h(\nabla\Phi_{\mathrm{on}}(h))^2 \\
&\quad + \chi_\perp\sigma^{-1}h\nabla\Phi_{\mathrm{on}}(h)\cdot\nabla\left(\Phi_{\mathrm{on}}'(h)\delta h\right) \\
&= \nabla\cdot\left[\chi_\perp\sigma^{-1}h\nabla\Phi_{\mathrm{on}}(h)\Phi_{\mathrm{on}}'(h)\delta h\right] \\
&\quad - \chi_\perp\sigma^{-1}h(\nabla^2\Phi_{\mathrm{on}}(h))\Phi_{\mathrm{on}}'(h)\delta h \\
&\quad + \frac{1}{2}\chi_\perp\sigma^{-1}(\nabla\Phi_{\mathrm{on}}(h))^2\delta h
\end{aligned}
\qquad (S11)
$$

to derive eq. (S9). We here approximately treat the nucleosome occupancy as a function of the brush height h to derive eq. (S11). This treatment is slightly different from the derivation of eq. (S5), where the nucleosome occupancy n_{his} is treated as independent of the brush height h.

We treat the 1d interface between domains of collapsed and swollen phases and thus the brush height is a function of the position x across the interface. Multiplying $dh(x)/dx$ to

both sides of eq. (S9) leads to the form

$$\frac{d}{dx}\left[-\frac{1}{2}Kh'^2(x) - \frac{1}{2}\chi_\perp\sigma^{-1}h\Phi_{\text{on}}'^2(h)h'^2(x) + \frac{W_\perp(h)}{T}\right] = 0. \tag{S12}$$

The work $W_\perp(h)$ has the form

$$W_\perp(h) = -\int_{h_{\text{swo}}}^{h} dh\, \Pi_\perp(h), \tag{S13}$$

where h_{swo} is the brush height in the bulk of the domains of the swollen phase. Eq. (S12) is effective across the interface, including the bulk of the swollen and collapsed domains. This leads to the form

$$\begin{aligned} -\frac{1}{2}Kh'^2(x) - \frac{1}{2}\chi_\perp\sigma^{-1}h\Phi_{\text{on}}'^2(h)h'^2(x) + \frac{W_\perp(h)}{T} &= \frac{W_\perp(h_{\text{swo}})}{T} \\ &= \frac{W_\perp(h_{\text{col}})}{T}, \end{aligned} \tag{S14}$$

where h_{col} is the brush height in the bulk of the domains of collapsed phase, because the brush height is uniform in the bulk of domains. The last equality of eq. (S14) leads to the Maxwell construction in the form

$$\int_{h_{\text{swo}}}^{h_{\text{col}}} dh\, \Pi_\perp(h) = 0. \tag{S15}$$

Although we here take into account the elastic energy of the grafting surface and the standard gradient energy to treat the interface, one can show that the Maxwell construction is effective for cases in which the free energy has other types of gradient terms as long as these gradient terms do not change the form of the first integral of the force balance equation in the bulk of domains.

S2 Chemical potentials and lateral osmotic pressure of DNA chains

In the layer structure, a part of swollen chains is in the interstitial region between collapsed chains (indicated by the subscript 'c') and the other part of swollen chains diffuses in the region above the collapsed chains (indicated by the subscript 's'). For cases in which the dynamics of DNA chain conformation is faster than the transcription dynamics and the dynamics of nucleosome assembly, the partition of swollen chains between these two regions is determined by the equality of chemical potentials, see fig. S1. The free energy density (per unit area) of swollen chains in the interstitial region has the form

$$\frac{f_c}{T} = \frac{3}{2}\frac{\sigma h_c^2}{N_c l_{\text{eff}}^2} + \frac{1}{2}w\frac{\sigma^2 N_c^2}{h_c}(n_{\text{his}} - n_+)(n_{\text{his}} - n_-) + \frac{1}{3}u\frac{\sigma^3 N_c^3}{h_c^2} + \frac{\Pi_{\text{app}}}{T}h_c, \qquad (S16)$$

where N_c is the number of chain segments in the interstitial region and h_c is the height of the collapsed chains, see also eq. (S1). The chemical potential of these chains thus has the form

$$
\begin{aligned}
\frac{\mu_c}{T} &= \sigma^{-1}\frac{\partial}{\partial N_c}\left(\frac{f_c}{T}\right) \\
&= -\frac{3}{2}\frac{h_c^2}{N_c^2 l_{\text{eff}}^2} + w\frac{\sigma N_c}{h_c}(n_{\text{his}} - n_+)(n_{\text{his}} - n_-) + u\frac{N_c^2\sigma^2}{h_c^2}. \qquad (S17)
\end{aligned}
$$

The free energy density (per unit area) of swollen chains in the region above collapsed chains has the form

$$
\begin{aligned}
\frac{f_s}{T} &= \frac{3}{2}\frac{\sigma\phi h_s^2}{N_s l_{\text{eff}}^2} + 3\frac{1-\phi}{N_s l_{\text{eff}}^2} + \frac{1}{2}w\frac{\sigma\phi^2 N_s^2}{h_s}(n_{\text{his}} - n_+)(n_{\text{his}} - n_-) + \frac{1}{3}u\frac{\sigma^3\phi^3 N_s^3}{h_s^2} \\
&\quad + \frac{\Pi_{\text{app}}}{T}h_s, \qquad (S18)
\end{aligned}
$$

see also eq. (S1). h_s is the height of the region above the collapsed chain and N_s $(\equiv 1 - N_c)$

is the number of chain segments in this region. The area occupied by swollen chains in this region is $(\phi\sigma)^{-1}$ per chain and may be comparable to (the square of) the radius of gyration of these chains, for cases in which the fraction ϕ of swollen chains is relatively small, see fig. S1. We thus take into account the second term of eq. (S18), which is the entropic elasticity with respect to the lateral excursions of swollen chains (to the region above collapsed chains). This term is derived by using the fact that the free energy due to the chain conformational entropy in the lateral direction (that is 2d) is $\frac{3k_BT}{Nl_{\text{eff}}^2}R^2$ per chain and the area R^2 occupied by a chain is confined to $(\phi\sigma)^{-1}$ due to the repulsive interactions with the neighboring swollen chains. The factor $(1-\phi)$ has been added by hand to make sure that the second term in eq. (S18) vanishes in the limit of a classical polymer brush ($\phi = 1$) where the swollen chains cannot perform excursions in the lateral direction.

The chemical potential of swollen chains in the region above the swollen chains has the form

$$
\begin{aligned}
\frac{\mu_{\text{s}}}{T} &= \sigma^{-1}\phi^{-1}\frac{\partial}{\partial N_{\text{s}}}\left(\frac{f_{\text{s}}}{T}\right) \\
&= -\frac{3}{2}\frac{h_{\text{s}}^2}{N_{\text{s}}^2 l_{\text{eff}}^2} - 3\frac{\phi^{-1}-1}{\sigma N_{\text{s}}^2 l_{\text{eff}}^2} + w\frac{\sigma N_{\text{s}}\phi}{h_{\text{s}}}(n_{\text{his}}-n_+)(n_{\text{his}}-n_-) + u\frac{\sigma^2\phi^2 N_{\text{s}}^2}{h_{\text{s}}^2}. \quad \text{(S19)}
\end{aligned}
$$

The number N_{c} of chain segments in the interstitial region between collapsed chains is derived by the equality of the chemical potential of chain segments, $\mu_{\text{s}} = \mu_{\text{c}}$ (see eqs. (S17) and (S19)). The lateral osmotic pressure $\Pi_{\parallel}$ of swollen chains is derived in the form

$$
\begin{aligned}
\frac{\Pi_{\parallel}(\phi)h_{\text{s}}(\phi)}{T} &= -\phi^2\frac{\partial}{\partial\phi}\left(\frac{f_{\text{s}}}{\phi T}\right) \\
&= -\frac{3}{N_{\text{s}}l_{\text{eff}}^2} + \frac{1}{2}w\frac{\sigma^2\phi^2 N_{\text{s}}^2}{h_{\text{s}}}(n_{\text{his}}-n_+)(n_{\text{his}}-n_-) + \frac{2}{3}u\frac{\sigma^3\phi^3 N_{\text{s}}^3}{h_{\text{s}}^2} \\
&\quad - \frac{\Pi_{\text{app}}h_{\text{s}}}{T}. \quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad\quad \text{(S20)}
\end{aligned}
$$

The minimum of the fraction ϕ of swollen chains is derived by the condition, $\Pi_{\parallel}(\phi) = 0$; for smaller values of the fraction ϕ, swollen chains cannot occupy the space above collapsed

chains and thus pressure Π_{app} is not applied to some of the collapsed chains (unless the grafting surface bends significantly).

The force balance equation of swollen chains has the form

$$\frac{\Pi_{\mathrm{app}}}{T} = -\frac{3\sigma\phi h_{\mathrm{s}}}{N_{\mathrm{s}}l_{\mathrm{eff}}^2} + \frac{1}{2}w\frac{\sigma^2\phi^2 N_{\mathrm{s}}^2}{h_{\mathrm{s}}^2}(n_{\mathrm{his}} - n_+)(n_{\mathrm{his}} - n_-) + \frac{2}{3}u\frac{\sigma^3\phi^3 N_{\mathrm{s}}^3}{h_{\mathrm{s}}^3}, \qquad (S21)$$

see also eq. (S6). Substituting eq. (S21) into eq. (S20) leads to the form

$$\tilde{\Pi}_{\parallel}(\phi) = \frac{N}{N_{\mathrm{s}}}\frac{1}{(1 - \gamma n_{\mathrm{his}})^2}\left[\phi'\tilde{h}_{\mathrm{s}}^2 - \frac{1}{\sigma h_{\mathrm{Alx}}^2}\right], \qquad (S22)$$

where we used rescaled height $\tilde{h}_{\mathrm{s}}$ ($\equiv h_{\mathrm{s}}/h_{\mathrm{Alx}}$) and rescaled lateral osmotic pressure $\tilde{\Pi}_{\parallel}(\phi)$ ($\equiv \Pi_{\parallel}(\phi)/\Pi_{\mathrm{Alx}}$). The second term in the square bracket is the contribution of the lateral entropic elasticity of swollen chains and the first term is the other contributions. The lateral entropic elasticity is thus negligible for cases in which the grafting density is very large, $\sigma h_{\mathrm{Alx}}^2 \gg 1$.

S3 Work done by the applied pressure

In the main article, we treat a process of i) applying pressure to a fraction $(1 - \phi)$ of chains in a uniformly swollen brush to collapse these chains and ii) let the other chains diffuse in the space above the collapsed chains. The work done by an external force in this process has the form

$$W(\phi) = W_{\perp}(\phi) + W_{\parallel}(\phi). \qquad (S23)$$

The work $W_{\perp}(\phi)$ is done in the process i) and has the form

$$W_{\perp}(\phi) = -(1 - \phi)A\int_{h_{\mathrm{s}}^* + h_{\mathrm{c}}}^{h_{\mathrm{c}}} dh\,\Pi_{\perp}(h). \qquad (S24)$$

The work $W_\parallel(\phi)$ is done in the process ii) and has the form

$$W_\parallel(\phi) = -\int_{\phi A}^{A} dA' \, \Pi_\parallel(\phi)h_{\mathrm{s}}(\phi) + \int_{h_{\mathrm{s}}^*}^{h_{\mathrm{s}}(\phi)} dh'_{\mathrm{s}}\frac{\Pi_{\mathrm{app}}}{T}A'. \qquad (\mathrm{S25})$$

The first term of eq. (S25) is the work done by the lateral osmotic pressure $\Pi_\parallel(\phi)$ in the process ii) and the second term is the work done by the applied pressure Π_{app} because the height $h_{\mathrm{s}}(\phi) + h_{\mathrm{c}}$ of the swollen chains changes in this process. A' in eq. (S25) is the area occupied by swollen chains. Transforming the integration variable A' to $\phi A/\phi'$ leads to the form

$$\begin{aligned}
W_\parallel(\phi) &= A\phi \int_1^\phi \frac{d\phi'}{\phi'^2}\,\Pi_\parallel(\phi')h_{\mathrm{s}}(\phi') + A\phi\int_1^\phi \frac{d\phi'}{\phi'}\,\Pi_{\mathrm{app}}\frac{dh_{\mathrm{s}}(\phi')}{d\phi'}\\
&= A\phi\int_1^\phi \frac{d\phi'}{\phi'^2}\left(\Pi_\parallel(\phi') + \Pi_{\mathrm{app}}\right)h_{\mathrm{s}}(\phi') + A\Pi_{\mathrm{app}}\left(h_{\mathrm{s}}(\phi) - \phi h_{\mathrm{s}}^*\right). \qquad (\mathrm{S26})
\end{aligned}$$

S4 Numerical calculations

We here summarize the details of the numerical calculations that were performed to derive fig. 6 in the main article. The other figures were derived in a similar manner. The work $W(\phi)$ that is necessary to change a uniform swollen brush to a layered brush is calculated by using the procedure shown in sec. 2 in the main article. We derive the fraction ϕ of swollen chains by numerically finding the global minimum of the work $W(\phi)$ (see also eq. (17) in the main article).

When the rescaled rate constant η_0 (defined by eq. (22) in the main article) is smaller than a threshold value $\eta_{0\mathrm{TP}}$, the fraction ϕ at the global minimum of the work is unity for any values of applied pressure (see fig. S3a). The work $W(\phi)$ shows another minimum for relatively large values of the rescaled rate constant η_0. The value of the work at this minimum is larger than zero as long as the rescaled rate constant η_0 is smaller than the threshold value $\eta_{0\mathrm{TP}}$. This implies that the layer structure is not stable for these values of the rescaled

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rate constant η_0. The DNA brush rather shows lateral phase separation. We thus used the Maxwell construction to derive the binodal curve that predicts the phase separation (see the green curve in fig. 6). This construction ensures that the work that is necessary to change a uniform swollen brush to a uniform collapsed brush is zero at the two-phase coexistent state.

For the rescaled rate constant η_0 that is larger than the threshold value $\eta_{0\text{TP}}$, the work at the new minimum becomes zero at a threshold applied pressure Π_{th1} (see fig. S3**b**). The work $W(\phi)$ at the minimum decreases with increasing applied pressure. Eventually, it becomes equal to the work to change a uniform swollen brush to a collapsed brush at another threshold applied pressure Π_{th2}. This implies that the layer structure is stable for applied pressures that are larger than the first threshold pressure Π_{th1} and smaller than the second threshold pressure Π_{th2}. These threshold pressures are shown by the blue curve in fig. 6 in the main article. These two threshold pressures and the binodal curve (for small values of the rescaled rate constant) intersect at the threshold rescaled rate constant $\eta_{0\text{TP}}$. This intersection is the triple point, at which a uniform swollen brush, a uniform collapsed brush, and the layer structure are all stable.

The fraction ϕ of swollen chains at the minimum of the work $W(\phi)$ for $\Pi_{\text{app}} = \Pi_{\text{th1}}$ (where the minimum of the work $W(\phi)$ becomes zero, see above) increases with increasing the rescaled rate constant η_0. It becomes unity at another threshold value $\eta_{0\text{tri}}$ of the rescaled rate constant (see the green curve in fig. S3**c**). At the threshold rate constant, the first and second derivative of the work $W(\phi)$ becomes zero at $\phi = 1$ for the first threshold pressure Π_{th1}. Indeed, this is the tricritical point. For a rescaled rate constant η_0 that is larger than the tricritical value, the minimum of the work is at $\phi = 1$ for the first threshold pressure Π_{th1} (see fig. S3**d**). The fraction ϕ at the minimum continuously decreases from unity with increasing applied pressure.

S5 Interfaces between swollen and collapsed chains

The interactions between chain segments and the entropy with respect to the lateral excursion of chains are modulated at the interfaces between swollen chains and collapsed chains. One may think that these interfacial effects destabilize the layer structure, which includes a rather large number of interfaces. We here show how the modulation of interactions and entropy at interfaces changes the phase diagram of DNA brushes. For simplicity, we here neglect the entropic elasticity with respect to lateral excursions of swollen chains (see the first term of eqs (16) and the second term of eq. (19) in the main article). We take into account the interfacial effects by using a simple form of excess work

$$W_{\text{int}}(\phi) = \chi_\perp h_c \sigma^{1/2} A \phi (1 - \phi) + \chi_\parallel (1 - \phi) \sigma A, \qquad (S27)$$

which is necessary to make the interfaces.

The first term of eq. (S27) is the work to make the vertical interfaces between collapsed chains and swollen chains at the interstitial region and the second term is the work to make the lateral interfaces between collapsed chains and swollen chains at the space above the collapsed chains. With eq. (S27), we treat the work to make a unit area of interfaces as constant. The constants $\chi_\perp$ and $\chi_\parallel$ account for the work to make vertical and lateral interfaces, respectively, and play an analogous role in interfacial tensions. The number of vertical interfaces is proportional to $\sigma A \phi (1 - \phi)$, see ref.[1] The number of lateral interfaces is proportional to the area $\sigma A (1 - \phi)$ occupied by the collapsed chains because we treat cases in which the space above these chains is occupied by swollen chains. To reduce the number of parameters, we use rescaled constants $\tilde{\chi}_\perp$ ($\equiv \sigma^{1/2} \chi_\perp / (\Pi_{\text{Alx}} h_{\text{Alx}})$) and $\tilde{\chi}_\parallel$ ($\equiv \sigma \chi_\parallel / \Pi_{\text{Alx}}$) in the following.

We first treat cases in which the (rescaled) vertical interfacial tension $\tilde{\chi}_\perp$ is very small. When the (rescaled) lateral interfacial tension $\tilde{\chi}_\parallel$ is relatively large, the phase diagram of a DNA brush has features that are qualitatively similar to cases in which the lateral entropic

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elasticity is significant (see fig. S6**a**). The values of the rescaled rate constant η_0 at the triple point and tricritical point increase with increasing the lateral interfacial tension $\tilde{\chi}_\parallel$ (see the solid curves in S6**b**), just like decreasing the grafting density (see fig. 8 in the main article). This is because the lateral interfacial tension $\tilde{\chi}_\parallel$ suppresses the lateral osmotic pressure of swollen chains, just like the lateral entropic elasticity, although the functional forms of the work due to these contributions are different. This implies that the fact that DNA brushes show tricritical points is relatively generic and is not very sensitive to the type of contributions (and thus the details of the functional form) that oppose the lateral osmotic pressure of swollen chains.

The DNA brush shows a tricritical point for a value of the lateral interfacial tension $\tilde{\chi}_\parallel$ that is larger than a threshold value (see fig. S6**b**). The threshold lateral tension $\tilde{\chi}_\parallel$ decreases with increasing the vertical interfacial tension $\tilde{\chi}_\perp$ and eventually becomes zero at a threshold value of the vertical tension $\tilde{\chi}_\perp$. For cases in which the lateral tension $\tilde{\chi}_\parallel$ is zero, the phase diagram of the DNA brush is qualitatively similar to cases in which the lateral entropic elasticity is negligible, as long as the vertical tension $\tilde{\chi}_\perp$ is smaller than the threshold value. For vertical tensions $\tilde{\chi}_\perp$ larger than the threshold value, the DNA brush shows a tricritical point at a value $\eta_{0\text{tri}}$ of the rescaled rate constant, but neither shows triple points nor lateral phase separation (see fig. S7**a** and **b**). This situation is very different from cases in which the lateral tension $\tilde{\chi}_\parallel$ is relatively large. For rescaled rate constants η_0 larger than the critical value $\eta_{0\text{tri}}$, the fraction ϕ of swollen chains continuously changes from unity at the first threshold pressure Π_{th1} (see also fig. S8). This results from the fact that the vertical interfacial tension does not suppress the lateral osmotic pressure. It rather increases the work that is necessary to collapse chains and thus tends to increase the fraction ϕ of swollen chains.

References

(1) Safran, S. A. *Statistical Thermodynamics of Surfaces, Interfaces, and Membranes* (Westview, 2003).
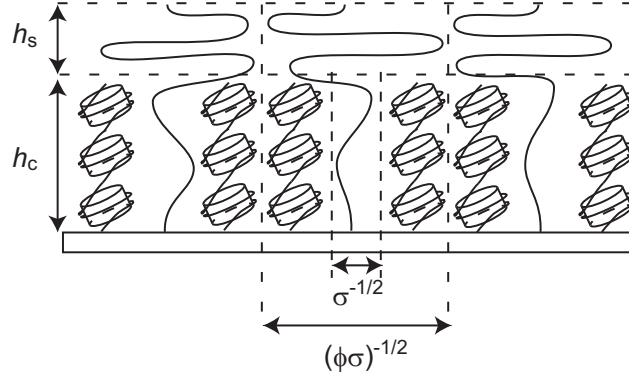
Figure S1: Segments of swollen chains are partitioned into the interstitial region between collapsed chains (this region is indicated by the subscript 'c') and the region above the collapsed chains (this region is indicated by the subscript 's'). h_s and h_c are the heights of the corresponding regions. N_s and N_c are the number of segments in these regions ($N_s + N_c = N$).
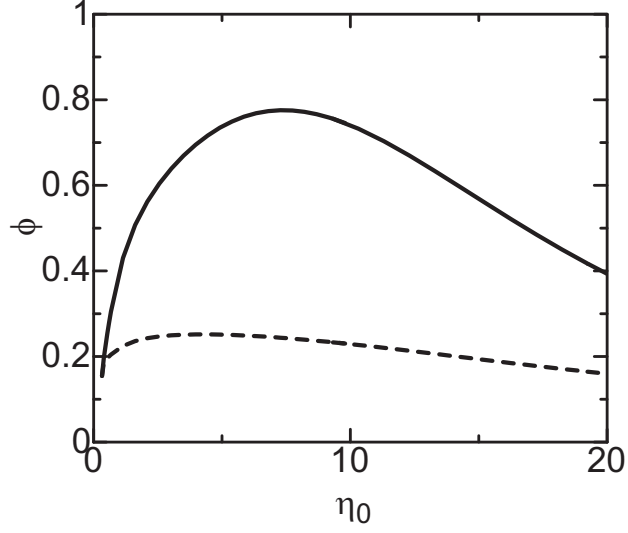
Figure S2: The fraction ϕ of swollen chains at $\Pi_{\mathrm{app}} = \Pi_1$ (solid) and Π_2 (broken) is shown as functions of the rescaled rate constant η_0 for cases in which the grafting density is very large, $\sigma h_{\mathrm{Alx}}^2 \gg 1$. The value of ϕ is maximum at $\Pi_{\mathrm{app}} = \Pi_1$ and is minimum at $\Pi_{\mathrm{app}} = \Pi_2$. The values of parameters that are used for the calculations are $n_+ = 0.99$, $n_- = -0.1$, $4uN\sigma/(3|w|h_{\mathrm{Alx}}) = 0.002$, $v\sigma N/h_{\mathrm{Alx}} = 0.8$, and $\gamma = 0.7$.
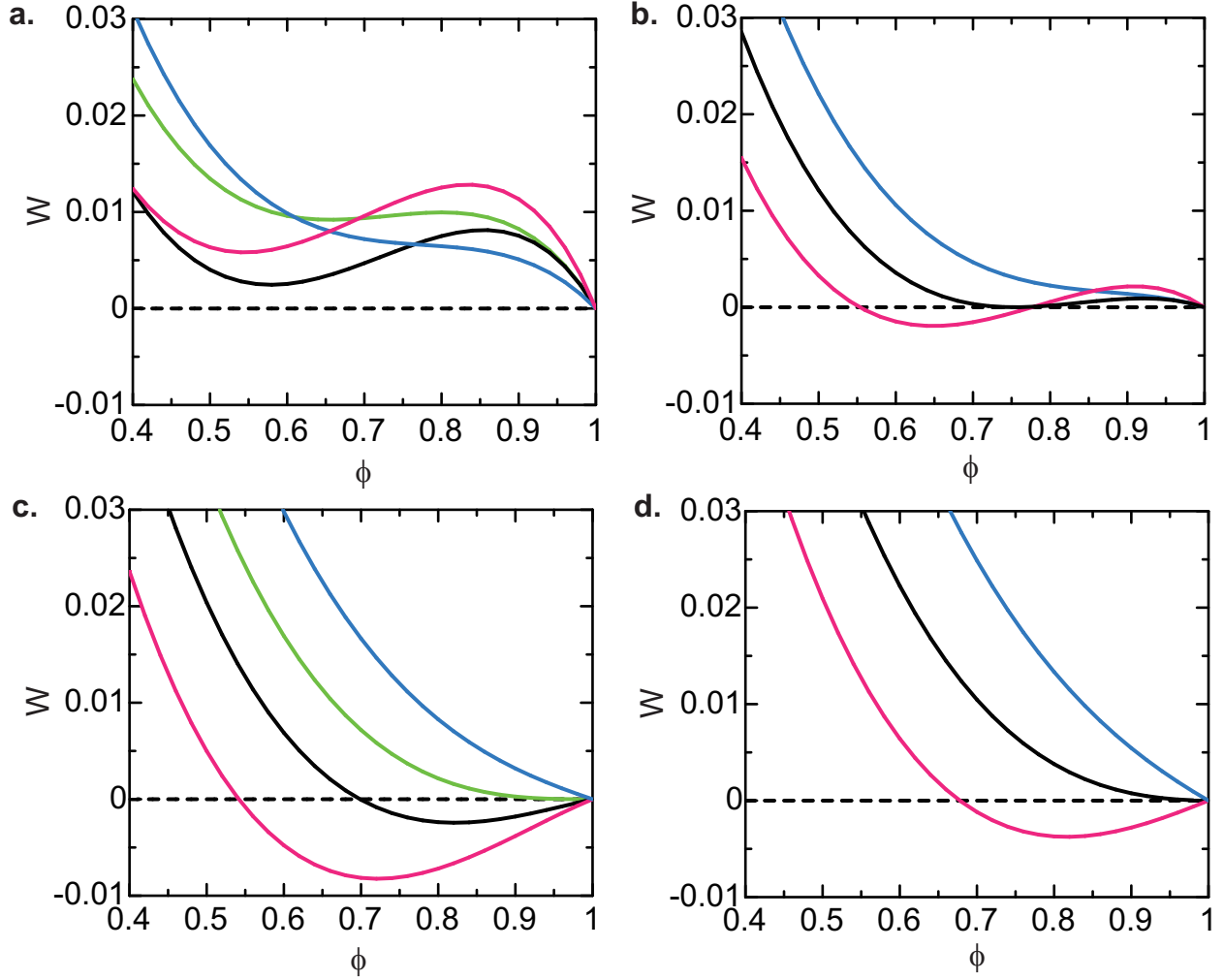
Figure S3: The work $W(\phi)$ that is necessary to change a uniform swollen brush to a layered brush is shown as functions of the fraction ϕ of swollen chains for cases in which the grafting density is relatively small ($\sigma h_{\mathrm{Alx}}^2 = 10.0$). The values of the rescaled rate constant η_0 are 1.55 (**a**), 1.62 (**b**), 1.73 (**c**), and 1.8 (**d**). The values of parameters that are used for the calculations are $n_+ = 0.99$, $n_- = -0.1$, $4uN\sigma/(3|w|h_{\mathrm{Alx}}) = 0.002$, $v\sigma N/h_{\mathrm{Alx}} = 0.8$, $\gamma = 0.7$, and $\sigma h_{\mathrm{Alx}}^2 = 10.0$. Color codes indicate the calculations for different values of applied pressure. Applied pressure increases in the order of cyan, black, light green, and magenta: **a.** $\Pi_{\mathrm{app}}/\Pi_{\mathrm{Alx}} = 3.5$ (cyan), 3.7 (light green), 3.9 (black), 4.0 (magenta), **b.** $\Pi_{\mathrm{app}}/\Pi_{\mathrm{Alx}} = 3.3$ (cyan), 3.469 (black), 3.7 (magenta), **c.** $\Pi_{\mathrm{app}}/\Pi_{\mathrm{Alx}} = 3.0$ (cyan), 3.15764 (light green), 3.3 (black), 3.5 (magenta), **d.** $\Pi_{\mathrm{app}}/\Pi_{\mathrm{Alx}} = 2.9$ (cyan), 3.1 (black), 3.3 (magenta).
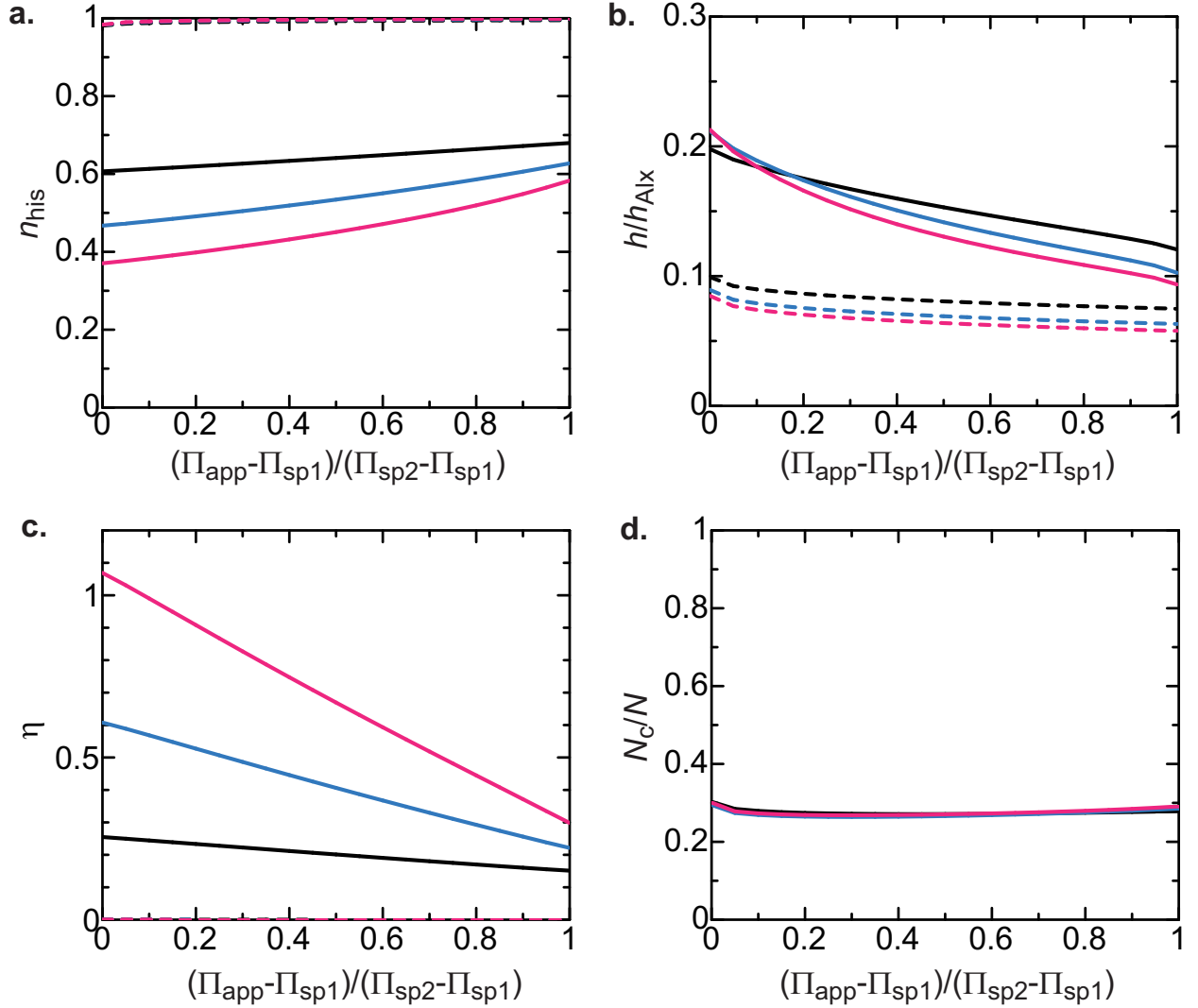
Figure S4: The nucleosome occupancy n_{his} (**a**), the brush height h/h_{Alx} (**b**), the rescaled transcription rate η (**c**), and the partition N_c/N of the segments of swollen chains to the interstitial region between collapsed chains (**d**) are shown as functions of applied pressure for cases in which the grafting density is very large $\sigma h_{Alx}^2 \gg 1$. Applied pressure is rescaled by the values, Π_1 and Π_2, at the spinodal curves. The values of swollen chains are shown by the solid curves and the values of collapsed chains are shown by the broken curve. The values of the rescaled rate constant that are used for the calculations are $\eta_0 = 1.0$ (black), 2.0 (cyan), 3.0 (magenta). The values of parameters that are used for the calculations are $n_+ = 0.99$, $n_- = -0.1$, $4uN\sigma/(3|w|h_{Alx}) = 0.002$, $v\sigma N/h_{Alx} = 0.8$, and $\gamma = 0.7$. These values of parameters correspond to fig. 5 in the main article.

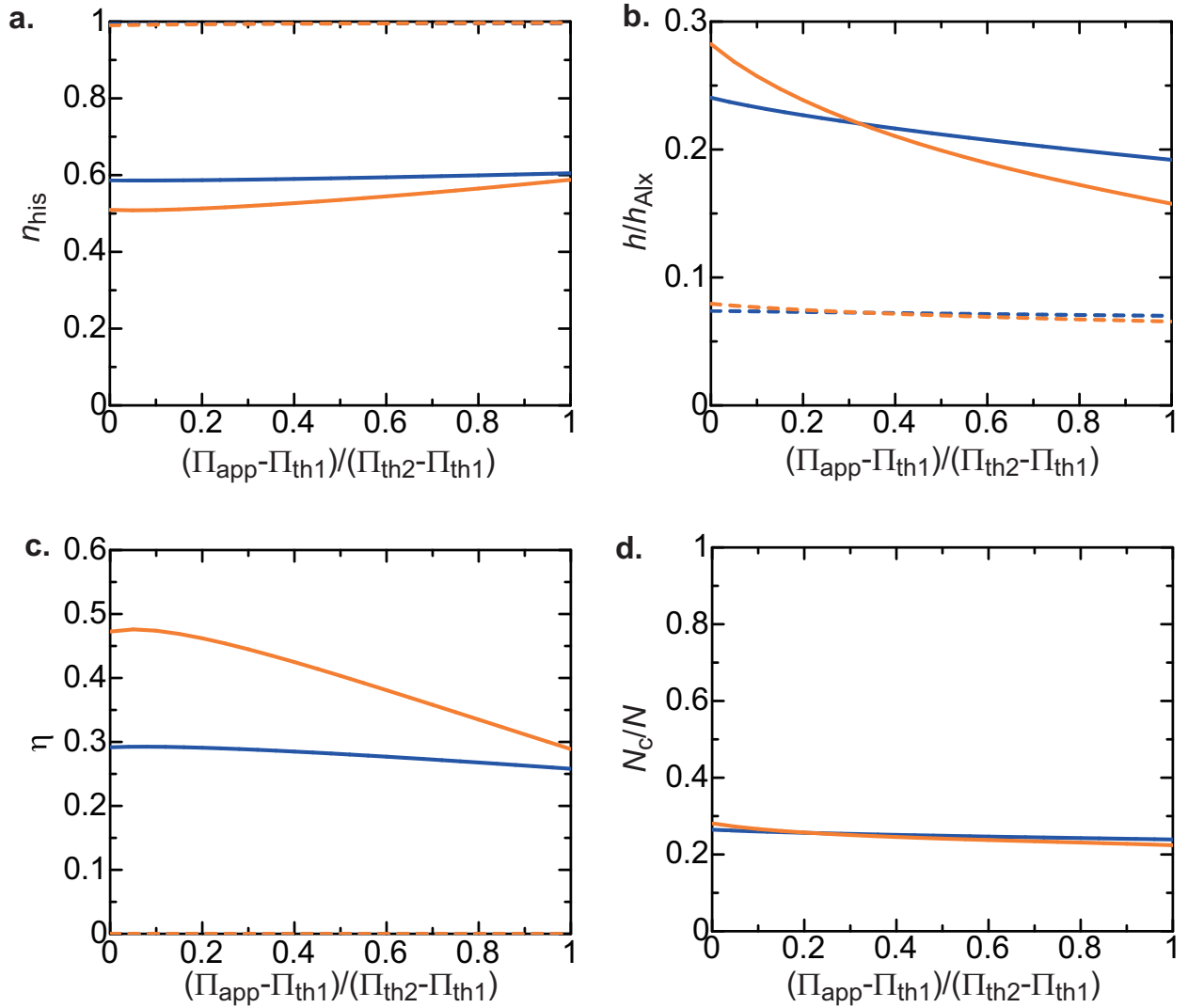Figure S5: The nucleosome occupancy n_{his} (**a**), the brush height h/h_{Alx} (**b**), the rescaled transcription rate η (**c**), and the partition N_{c}/N of the segments of swollen chains to the interstitial region between collapsed chains (**d**) are shown as functions of applied pressure for cases in which the grafting density is relatively small $\sigma h_{\mathrm{Alx}}^2 = 10.0$. Applied pressure is rescaled by the values, Π_{th1} and Π_{th2}, at the binodal curves. The values of swollen chains are shown by the solid curves and the values of collapsed chains are shown by the broken curve. The values of the rescaled rate constant that are used for the calculations are $\eta_0 = 1.65$ (blue) and 2.0 (orange). The values of parameters that are used for the calculations are $n_+ = 0.99$, $n_- = -0.1$, $4uN\sigma/(3|w|h_{\mathrm{Alx}}) = 0.002$, $v\sigma N/h_{\mathrm{Alx}} = 0.8$, and $\gamma = 0.7$. These values of parameters correspond to fig. 7 in the main article.
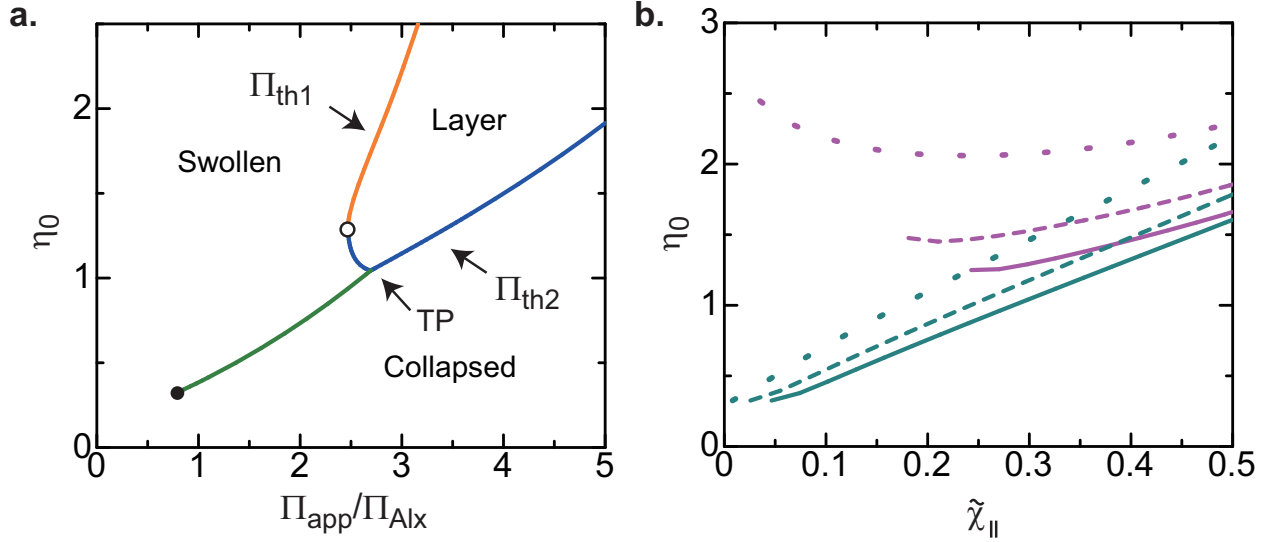
Figure S6: **a.** The phase diagram of a DNA brush is shown as a function of the rescaled rate constant η_0 and applied pressure $\Pi_{\mathrm{app}}/\Pi_{\mathrm{Alx}}$ for $\tilde{\chi}_\perp = 0$ and $\tilde{\chi}_\parallel = 0.3$ (see the discussion below eq. (S27) for the definition of $\tilde{\chi}_\perp$ and $\tilde{\chi}_\parallel$). The green line shows the applied pressure, at which the DNA brush shows lateral phase separation. The blue and orange lines delineate the region, in which the layer structure is stable. The brush shows first order phase transitions (with respect to the fraction ϕ of swollen chains) at the blue line and second order phase transitions at the orange lines. The filled circle shows the critical point and the unfilled circle shows the tricritical point. The intersection between the green and blue curves is the triple point ('TP'). **b.** The values of rescaled rate constant η_0 at the triple point (emerald green) and tricritical point (purple) are shown as functions of the constant $\tilde{\chi}_\parallel$ for $\tilde{\chi}_\perp = 0.0$ (solid), 1.0 (broken), and 3.0 (dotted). The other parameter values are $n_+ = 0.99$, $n_- = -0.1$, $\tilde{u} = 0.002$, $\tilde{v} = 0.8$, and $\gamma = 0.7$.
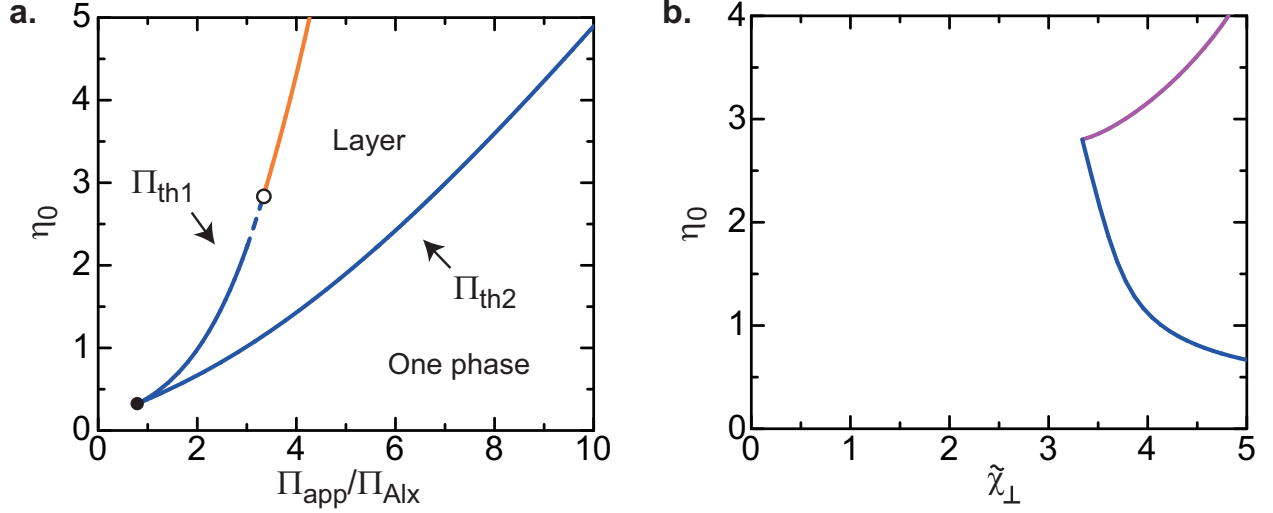
Figure S7: **a.** The phase diagram of a DNA brush is shown as a function of the rescaled rate constant η_0 and applied pressure $\Pi_{\mathrm{app}}/\Pi_{\mathrm{Alx}}$ for $\tilde{\chi}_\perp = 3.5$ and $\tilde{\chi}_\parallel = 0.0$ (see the discussion below eq. (S27) for the definition of $\tilde{\chi}_\perp$ and $\tilde{\chi}_\parallel$). The blue and orange lines delineate the region, at which the DNA brush shows the layer structure. The blue and orange lines indicate first order phase transition and second order phase transition (with respect to the fraction ϕ of swollen chains), respectively. The threshold applied pressure, Π_{th1} and Π_{th2}, is equal to the spinodal pressure, Π_{sp1} and Π_{sp2}, at the blue solid line and is larger than the spinodal pressure at the blue broken line or the orange line. The critical point is shown by the filled circle and the tricritical point is shown by the unfilled circle. **b.** The values of the rescaled rate constant η_0 at the tricritical point (purple) is shown as a function of the constant $\tilde{\chi}_\perp$ for $\tilde{\chi}_\parallel = 0$. The blue curve shows the values of the rescaled rate constant η_0 at the intersection between the blue solid and broken lines in **a**. The values of parameters that are used for the calculations are $n_+ = 0.99$, $n_- = -0.1$, $\tilde{u} = 0.002$, $\tilde{v} = 0.8$, and $\gamma = 0.7$.
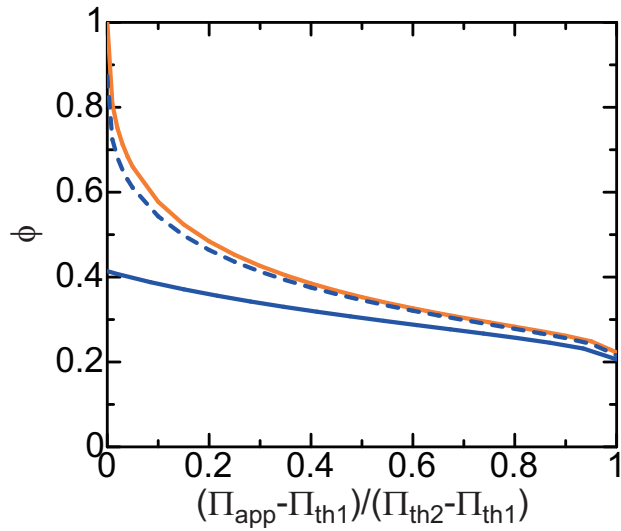
Figure S8: The fraction ϕ of swollen chains is shown as a function of applied pressure Π_{app} (rescaled by the threshold pressure Π_{th1} and Π_{th2}) for cases in which the interfacial tensions are $\tilde{\chi}_\perp = 3.5$ and $\tilde{\chi}_\parallel = 0$. The values of the rescaled rate constant η_0 are $\eta_0 = 2.0$ (blue solid), 2.5 (blue broken), and 3.0 (orange). The values of parameters that are used for the calculations are $n_+ = 0.99$, $n_- = -0.1$, $4uN\sigma/(3|w|h_{Alx}) = 0.002$, $v\sigma N/h_{Alx} = 0.8$, and $\gamma = 0.7$. These values correspond to fig. 10a in the main article.