Supporting Information: Transcription driven phase separation in chromatin brush

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S1 Formal formulation of the theory

S1.1 Uni-directional motion of RNAP along DNA chains

We here describe the formal formulation of our model in a self-contained manner. Some part of this section thus duplicates with the description in sec. 2 of the main article. Transcription is initiated when an RNAP binds to a non-coding DNA sequence, called promoter, by specific interactions and changes its conformation. The enzyme then moves uni-directionally towards another non-coding sequence, called terminator, base-by-base, while synthesizing a chain of RNA. When the RNAP reaches the terminator, it is released from the DNA. We treat cases where each DNA chain has a promoter at its free end and a terminator at its grafted end.

For the case that the occupancy of RNAP on DNA chain segments is relatively small (and thus that the RNAPs do not produce a traffic jam on the DNA molecules), the dynamics of the occupancy $n_{\rm rnp}(s_\beta, t)$ of RNAP is given by the following rate equations:

$$\begin{aligned}
\frac{\partial}{\partial t} n_{\rm rnp}(s_{\rm p}, t) &= \lambda \rho(h, t)(1 - n_{\rm rnp}(s_{\rm p}, t)) \\
&\quad -\xi n_{\rm rnp}(s_{\rm p}, t)(1 - n_{\rm his}(s_1, t)), && \text{(S1)} \\
\frac{\partial}{\partial t} n_{\rm rnp}(s_\beta, t) &= \xi n_{\rm rnp}(s_{\beta-1}, t)(1 - n_{\rm his}(s_\beta, t)) \\
&\quad -\xi n_{\rm rnp}(s_\beta, t)(1 - n_{\rm his}(s_{\beta+1}, t)), && \text{(S2)} \\
\frac{\partial}{\partial t} n_{\rm rnp}(s_{\rm t}, t) &= \xi n_{\rm rnp}(s_{N_0-1}, t) - k_{\rm off}^{\rm T} n_{\rm rnp}(s_{\rm t}, t). && \text{(S3)}
\end{aligned}$$

We here assume that the promoter and terminator regions are not occupied by nucleosomes due to the specific sequence of the promoters and terminators. s_β labels the binding sites of a DNA chain from the free end to the grafted end and β runs from 1 to $N_0 - 1$. $s_{\rm p}$ $(= s_0)$ and $s_{\rm t}$ $(= s_{N_0})$ indicate the promoter and the terminator, respectively.

The first term of eq. (S1) is the binding rate of RNAP to the promoter due to specific interactions, where λ is the rate constant that accounts for the binding. $\rho(z, t)$ is the local concentration of RNAP in the solution of the brush region (which is derived by using eq.

(S5)) and h is the height of the brush, see fig. 1 in the main article. We neglected the dissociation rate of RNAP from the promoters because RNAP firmly grips DNA chains once it changes its conformation. The second term of eq. (S1) is the rate at which RNAP moves to the next binding site. The motion of RNAP is suppressed when the next binding site is already occupied by nucleosomes. $n_{\rm his}(s_\beta, t)$ is the occupancy of nucleosomes on the site s_β ($\beta = 1, 2, \cdots, N_0 - 1$). ξ is the rate constant that accounts for the motion of RNAP. Fueled by RNA polymerization, RNAP moves uni-directionally site-by-site towards the terminators with the same rate, see eq. (S2). RNAP is a processive motor and thus once RNAP binds to the promoter, it is not dissociated from the DNA molecule before it reaches the terminator. The second term of eq. (S3) is the dissociation rate of RNAP from the terminators, where the rate constant $k_{\rm off}^{\rm T}$ accounts for the dissociation.

S1.2 Assembly and dissociation of nucleosomes

The dynamics of the assembly of nuclesomes is treated by using a rate equation of the form

$$\begin{aligned} \frac{\partial}{\partial t} n_{\rm his}(s_\beta, t) &= k_{\rm on}^{\rm his} c(z_\beta, t)(1 - n_{\rm his}(s_\beta, t)) - k_{\rm off}^{\rm his} n_{\rm his}(s_\beta, t) \\ &\quad -\zeta n_{\rm rnp}(s_{\beta-1}, t) n_{\rm his}(s_\beta, t), \end{aligned} \tag{S4}$$

where s_β labels the binding sites of a DNA chain from the free end to the grafted end and β runs from 1 to $N_0 - 1$. The first term of eq. (S4) is the rate of the nucleosome assembly on the β-th DNA chain segment and $k_{\rm on}^{\rm his}$ is the rate constant that accounts for this process. z_β is the height of the β-th chain segment above the surface ($\beta = 1, 2, \cdots, N_0 - 1$), and $c(z, t)$ is the local concentration of histone proteins in the solution of the brush region. The second term of eq. (S4) accounts for nucleosome dissociation by thermal excitation with rate constant $k_{\rm off}^{\rm his}$. The spontaneous dissociation of nucleosomes due to thermal excitation is a very slow process because relatively large free energy costs ($> 15\ k_B T$) are necessary to dissociate nucleosomes even when the dissociation is not suppressed by the attractive

interactions between nucleosomes. Henceforth, we thus neglect this process, $k_{\rm off}^{\rm his} \simeq 0$. The third term of eq. (S4) is the dissociation rate of nucleosomes due to the collision between nucleosomes and RNAP during transcription and ζ is the associated rate constant.

We neglected a couple of molecular details involved in the assembly of nucleosomes: i) the fact that nucleosomes are assembled from 8 histone proteins (and thus the assembly rate is $\sim c^8(z,t)$ in a more precise treatment), ii) the specific chemistry of four types of core histone proteins (H2A, H2B, H3, and H4), and iii) the fact that the assembly of nucleosomes is guided by chaperones. With the treatment ii), four different types of histone proteins are treated as one type of molecule by using the local density $c(z,t)$ for the cases that the solution includes H2A, H2B, H3, and H4 with equal concentrations.

S1.3 Diffusion of RNA polymerase in solutions

The local concentrations $\rho(z,t)$ of RNAP in the solution of the brush region are derived by using the diffusion equation that has the form

$$\begin{aligned} \frac{\partial}{\partial t}\rho(z,t) &= -\frac{\partial}{\partial z}J_{\rm rnp}(z,t) - \lambda\rho(z,t)g_{\rm p}(z,t) \\ &\quad +k_{\rm off}^{\rm rnp}(z,t), \end{aligned} \tag{S5}$$

where the first term is due to the flux $J_{\rm rnp}(z,t)$ of RNAP in the solution of the brush region, the second term accounts for the binding of RNAP to the promoters, and the third term for the unbinding of RNAP from the terminators. λ is the rate constant that accounts for the binding of RNAP to the promoters, see also eq. (S1). $g_{\rm p}(z)$, the local concentration of the promoters, is given by $g_{\rm p}(z) = \sigma\delta(z-h)$ because with the Alexander approximation, the promoters of all the DNA chains in the brush are located at the top of the brush. $k_{\rm off}^{\rm rnp}(z,t)$ is the rate constant that accounts for the release of RNAP from the terminators. There is no general form of this rate constant because the releasing rate of RNAP from the terminator of a DNA chain depends on the binding rate of RNAP at the promoters of the same chain

(see also sec. S1.6).

The flux $J_{\rm rnp}(z,t)$ has the form

$$J_{\rm rnp} = -D_{\rm rnp}\left[\frac{\partial}{\partial z}\rho(z,t) + v\rho(z,t)\frac{\partial}{\partial z}\Phi_{\rm on}(z)\right], \tag{S6}$$

where the first term is the flux due to the thermal diffusion and the second term is the flux due to the non-specific interactions between nucleosomes and RNAP. $D_{\rm rnp}$ is the diffusion constant of RNAP. $\Phi_{\rm on}(z)$ is the local concentrations of nucleosomes and v is the 2nd virial coefficient that accounts for the non-specific interactions between nucleosomes and RNAP in the solution. Without changing the physics, we neglect the non-specific interactions between vacant DNA chain segments and RNAP in the solution. We treat cases, in which the local concentration of RNAP in the solution of the brush region is very small and thus the interactions between RNAP molecules (and also the interactions between RNAP and histone proteins) are negligible.

S1.4 Diffusion of histone proteins in solutions

The local concentrations $c(z,t)$ of histones are derived by using the diffusion equation that has the form

$$\frac{\partial}{\partial t}c(z,t) = -\frac{\partial}{\partial z}J_{\rm his}(z,t) - k_{\rm on}^{\rm his}c(z,t)\Phi_{\rm off}(z,t) + S_{\rm off}(z,t), \tag{S7}$$

where the first term is due to the flux $J_{\rm his}(z,t)$ of histone proteins, the second term is the assembling rate of nucleosomes at the binding sites of DNA chains, and the third term is the dissociation rate of nucleosomes from the binding sites of DNA chains. $k_{\rm on}^{\rm his}$ is the rate constant that accounts for the assembly of nucleosomes and $\Phi_{\rm off}(z,t)$ is the local concentration of vacant DNA chain segments (which are not occupied by nucleosomes). The dissociation rate $S_{\rm off}(z,t)$ of nucleosomes has the form $\zeta\Phi_{\rm col}(z,t)$, where $\Phi_{\rm col}(z,t)$ is the local concentrations of nucleosomes that are colliding with RNAP during transcription, see also the third

term of eq. (S4).

The flux $J_{\rm his}(z,t)$ has the form

$$J_{\rm his}(z,t) = -D_{\rm his}\left[\frac{\partial}{\partial z}c(z,t) + v_{\rm his}c(z,t)\frac{\partial}{\partial z}\Phi_{\rm on}(z,t)\right]. \tag{S8}$$

The first term of eq. (S8) is the flux due to the thermal diffusion and the second term of this equation is the flux due to the non-specific interactions between nucleosomes and histone proteins in the solution. $D_{\rm his}$ is the diffusion constant of histone proteins in the solution of the brush region and $v_{\rm his}$ is the second virial coefficient that accounts for the non-specific interactions between nucleosomes and histone proteins in the solution. The size of histone proteins is much smaller than the size of RNAP and, henceforth, we thus neglect the interactions between histone proteins and nucleosomes (and also the non-specific interactions between histone proteins and vacant DNA chain segments). We treat the cases that the local concentration of histone in the solution of the brush region is relatively small and thus the interactions between histone proteins (and also the interactions between RNAP and histone) are negligible.

S1.5 Free energy of chromatin brush

For the cases that the dynamics of the conformation of DNA chains is faster than the other processes, the height h of a DNA brush is derived by minimizing the free energy (per unit area) that has the form

$$f = f_{\rm pol} + f_{\rm int} + \Pi_{\rm app}h \tag{S9}$$

with respect to the brush height h. $f_{\rm pol}$ is the free energy due to the entropic elasticity of DNA chains, $f_{\rm int}$ is the free energy due to non-specific interactions, and the third term is the work done by an applied pressure $\Pi_{\rm app}$. In this paper, we treat two cases: i) a DNA brush alone in a solution, $\Pi_{\rm app} = 0$, and ii) a DNA brush pushed against another DNA brush with

applied pressures $\Pi_{\rm app}$ (> 0), see fig. 1 in the main article. In case ii), the functional form of the free energy for the two brushes is identical (although the values of the free energy may be different in some cases) and thus the free energy of the system is the sum of the free energies of the two brushes.

The free energy $f_{\rm pol}$ due to the entropic elasticity of DNA chains has the form

$$\frac{f_{\rm pol}}{T} = \frac{3}{2}\frac{\sigma h^2}{N_0 l_{\rm eff}^2}, \tag{S10}$$

where T is the absolute temperature in units of the Boltzmann constant. We use the form $l_{\rm eff} = l_{\rm a}(1 - \gamma n_{\rm his}(s_\beta, t))$ to treat the effective length $l_{\rm eff}$ of chain segments ($\beta = 1,\ 2,\ \cdots,\ N_0 - 1$), where the constant $\gamma > 0$ accounts for the fact that the length of DNA chain segments becomes shorter when they are reeled around histone proteins.

The free energy $f_{\rm int}$ due to non-specific interactions has the form

$$\begin{aligned}\frac{f_{\rm int}}{T} &= \int dz\ \left[\frac{1}{2}w_{\rm on}\Phi_{\rm on}^2(z) + w_{\rm int}\Phi_{\rm on}(z)\Phi_{\rm off}(z) + \frac{1}{2}w_{\rm off}\Phi_{\rm off}^2(z)\right] \\ &\quad + \frac{1}{3}u\int dz\,\Phi_{\rm on}^3(z).\end{aligned} \tag{S11}$$

The 2nd virial coefficients $w_{\rm on}$, $w_{\rm int}$, and $w_{\rm off}$ account for the (nucleosome)-(nucleosome) interactions, the (nucleosome)-(vacant DNA segment) interactions, and the (vacant DNA segment)-(vacant DNA segment) interactions, respectively; the interactions between DNA chain segments change from repulsive to attractive when nucleosomes are assembled at the DNA chain segments. u is the 3rd virial coefficient that accounts for the three-body interactions between nucleosomes. We here treat the cases that the local concentrations of RNAP and histone are relatively small in the solution of the brush region and thus neglected the interactions between these proteins and DNA chain segments. For simplicity, we do not explicitly treat the fact that nucleosomes, due to their attractive interactions, might assemble into chromatin fibers.

S1.6 Steady states

Because we neglected the non-specific interactions between histone proteins and nucleosomes, the local concentrations $c(z,t)$ of histones are equal to the concentration c_0 of histones in the bulk solution. In steady states, the solutions of eqs. (S1) - (S4) have the forms

$$n_{\rm rnp} = \frac{\lambda\rho(h)}{2\xi}\left(1+\sqrt{1+\frac{4}{\eta}}\right) \tag{S12}$$

$$n_{\rm his} = \frac{1}{2}\eta\left(1+\frac{2}{\eta}-\sqrt{1+\frac{4}{\eta}}\right) \tag{S13}$$

with

$$\eta = \frac{\lambda\rho(h)\zeta}{k_{\rm on}^{\rm his}c_0\xi}. \tag{S14}$$

In the uniform DNA brush, the occupancies, $n_{\rm rnp}(s_\beta,t)$ and $n_{\rm his}(s_\beta,t)$, of RNAP and nucleosomes do not depend on the position of the binding sites s_β. For simplicity, we use $n_{\rm rnp}$ $(=n_{\rm rnp}(s_\beta,t))$ and $n_{\rm his}$ $(=n_{\rm his}(s_\beta,t))$ to represent these occupancies. Eq. (S13) implies that the local concentrations of nucleosomes and vacant DNA chain segments are uniform and have the forms

$$\Phi_{\rm on}(z) = \frac{\sigma N_0}{h}n_{\rm his} \tag{S15}$$

$$\Phi_{\rm off}(z) = \frac{\sigma N_0}{h}(1-n_{\rm his}). \tag{S16}$$

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In steady states, the binding rate of RNAP to the promoter of a DNA chain is equal to the dissociation rate of RNAP from the terminator of the chain, see eqs. (S1) - (S3). The rate constant $k_{\rm off}^{\rm rnp}(z)$ thus has the form $k_{\rm off}^{\rm rnp}(z)=\lambda\sigma\rho(h)\delta(z)/h$. We derive the solution of eq. (S5) by enforcing two boundary conditions: i) the local concentration $\rho(h)$ of RNAP at the top of the brush is determined by the continuity of the chemical potentials of RNAP,

$\rho(h) = \rho_0 \mathrm{e}^{-v\Phi_{\rm on}(h)}$, and ii) the flux of RNAP at the solid surface ($z = 0$) is equal to the rate of RNAP molecules that are released at the terminators, $J_{\rm rnp}(0) = \lambda\sigma\rho(h)/h$ (because RNAP cannot penetrate the solid surface). With these boundary conditions the local concentration of RNAP in the solution of the brush region has the form

$$\rho(z) = \rho(h)\left[1 + \frac{\lambda\sigma}{D_{\rm rnp}}(h - z)\right]. \tag{S17}$$

With boundary condition i) we assume that although the promoters of DNA chains are located at their free ends, the local concentration of RNAP at the vicinity of the promoters is smaller than the concentration of the bulk solution due to the excluded volume interactions between nucleosomes and RNAP. Experimentally, this may be effective for the cases that there is a small distance between the free ends and the promoters of DNA chains.

Minimizing eq. (S9) with respect to the brush height h leads to the force balance equation

$$\begin{aligned} -\frac{\Pi_{\rm app}}{T} &= \frac{3\sigma h}{N_0 l_{\rm eff}^2} - \frac{w\sigma^2 N_0^2}{2h^2}(n_{\rm his} - n_+)(n_{\rm his} - n_-) \\ &\quad -\frac{2}{3}u\frac{\sigma^3 N_0^3}{h^3}, \end{aligned} \tag{S18}$$

with $w = w_{\rm on} + w_{\rm off} - 2w_{\rm int}$ and $n_\pm = (w_{\rm off} - w_{\rm int} \pm \sqrt{w_{\rm int}^2 - w_{\rm on}w_{\rm off}})/w$. We derived the third term of eq. (S18) by using the fact that this term is only significant for $n_{\rm his} \simeq 1$. Indeed, eq. (S7) is automatically satisfied with the above solutions (with $\Phi_{\rm col} = \sigma N_0 n_{\rm his} n_{\rm rnp}/h$).

The brush height is derived as a function of the occupancy $n_{\rm his}$ of nucleosomes by using eq. (S18). The parameter η is thus a function of the occupancy $n_{\rm his}$ via $\rho(h)$, see eq. (S15). Eq. (S13) is thus a self-consistent equation of the occupancy $n_{\rm his}$. We use eq. (S13) to derive the occupancy of nucleosomes as functions of the rate constants that are relevant to transcription, the extent of the modulation of interactions between DNA chain segments by assembling nucleosomes, and applied pressures.

S2 Diffusion of RNAP in the solution

We here derive the fluxes of RNAP in the solution of the brush region, eq. (5) in the main article. It is an extension of the derivation that is shown in sec. S1 in the electronic supplementary information in ref.[1] The free energy contributions that are relevant to the diffusion of RNAP (which depend on the local concentration $\rho(z,t)$ of RNAP in the solution) have the form

$$\frac{f_{\rm rnp}}{T} = \int dz \left[\rho(z,t)(\log \rho(z,t) - 1) + v\rho(z,t)\Phi_{\rm on}(z,t)\right]. \tag{S19}$$

The first term of eq. (S19) is the free energy contributions due to the translational entropy of RNAP and the second term is the free energy contributions due to the interactions between RNAP and nucleosomes. T is the absolute temperature. v is the second virial coefficients that account for the interactions between RNAP and nucleosomes. $\Phi_{\rm on}(z,t)$ is the local concentration of nucleosomes. With eq. (S19), we neglected the (histone)-(RNAP) interactions, the (vacant DNA segment)-(RNAP) interactions, and the (RNAP)-(RNAP) interactions, see sec. 2 in the main article.

The chemical potential has the form

$$\begin{aligned} \frac{\mu_{\rm rnp}(z,t)}{T} &\equiv \frac{1}{T}\frac{\delta f_{\rm rnp}}{\delta \rho(z,t)} \\ &= \log \rho(z,t) + v\Phi_{\rm on}(z,t), \end{aligned} \tag{S20}$$

where $\delta f_{\rm rnp}/\delta\rho(z,t)$ is the functional derivative of the free energy $f_{\rm rnp}$ with respect to the local concentration $\rho(z,t)$. By using the Debye construction, the fluxes of RNAP in the solution of the brush region are derived in the form

$$\begin{aligned} J_{\rm rnp}(z,t) &\equiv -D_{\rm rnp}\rho(z,t)\frac{\partial}{\partial z}\frac{\mu_{\rm rnp}(z,t)}{T} \\ &= -D_{\rm rnp}\left[\frac{\partial}{\partial z}\rho(z,t) + v\rho(z,t)\frac{\partial}{\partial z}\Phi_{\rm on}(z,t)\right], \end{aligned} \tag{S21}$$

where we used the Einstein's relationship to derive the last form of eq. (S21). The last form of eq. (S21) is equal to eq. (5) in the main article.

The chemical potential of RNAP in the bulk solution has the form

$$\frac{\mu_0}{T} = \log \rho_0, \tag{S22}$$

where ρ_0 is the concentration of RNAP in the bulk solution. The continuity of chemical potentials at the interface between the brush region and the bulk solution, $\mu_{\rm rnp}(h,t) = \mu_0$, lead to the local concentration $\rho(h)$ of RNAP at the top of the brush in the form

$$\rho(h) = \rho_0 \mathrm{e}^{-v\Phi_{\rm on}(h)}. \tag{S23}$$

Eq. (S23) is equal to eq. (7) in the main article.

S3 Linear stability analysis

To analyze the stability of the steady state solutions, we consider small fluctuations

$$\tilde{n}_{\rm his} = n_{\rm his} + \delta n_{\rm his} \tag{S24}$$

$$\tilde{n}_{\rm rnp} = n_{\rm rnp} + \delta n_{\rm rnp} \tag{S25}$$

around the steady state solutions, $n_{\rm his}$ and $n_{\rm rnp}$, see eqs. (16) and (17) in the main article. Substituting eqs. (S24) and (S25) into eqs. (S1) and (S4) and expanding these equations in the power series of $\delta n_{\rm his}$ and $\delta n_{\rm rnp}$ lead to the form

$$\frac{\partial}{\partial t}\begin{pmatrix} \delta n_{\rm his} \\ \delta n_{\rm rnp} \end{pmatrix} = -\begin{pmatrix} k_{\rm on}^{\rm his} c_0 + \zeta n_{\rm rnp} & \zeta n_{\rm his} \\ -\left(\xi n_{\rm rnp} + \lambda \frac{d\rho(h)}{dn_{\rm his}}\right) & \xi(1-n_{\rm his}) \end{pmatrix}\begin{pmatrix} \delta n_{\rm his} \\ \delta n_{\rm rnp} \end{pmatrix}. \tag{S26}$$

Because the diagonal elements of the coefficient matrix of eq. (S26) are positive, the steady state solutions, $n_{\rm his}$ and $n_{\rm rnp}$, are stable when the determinant of this matrix

$$\begin{aligned}\Delta &= \xi(1-n_{\rm his})(k_{\rm on}^{\rm his}c_0+\zeta n_{\rm rnp})+\zeta n_{\rm his}\left(\xi n_{\rm rnp}+\lambda\frac{d\rho(h)}{dn_{\rm his}}\right)\\ &= \xi k_{\rm on}^{\rm his}c_0\eta\sqrt{1+\frac{4}{\eta}}\left(1-\frac{d}{dn_{\rm his}}g(\eta)\right)\end{aligned} \tag{S27}$$

is positive. The function $g(\eta)$ is the right hand side of eq. (17) in the main article;

$$g(\eta)=\frac{1}{2}\eta\left(1+\frac{2}{\eta}-\sqrt{1+\frac{4}{\eta}}\right). \tag{S28}$$

Eq. (17) in the main article has maximal three solutions, $n_{\rm his}^{(1)}$, $n_{\rm his}^{(2)}$, and $n_{\rm his}^{(3)}$ ($n_{\rm his}^{(1)} < n_{\rm his}^{(2)} < n_{\rm his}^{(3)}$). The above argument suggests that two of the solutions, $n_{\rm his}^{(1)}$ and $n_{\rm his}^{(3)}$, are stable, whereas the other solution $n_{\rm his}^{(2)}$ is unstable. One of the stable solutions become unstable when the solution satisfies a condition

$$\frac{d}{dn_{\rm his}}g(\eta)=1, \tag{S29}$$

see eq. (S27), where the solution becomes equal to the unstable solution, analogous to the spinodal curve of the usual case of phase separation. We thus derive the spinodal curve in fig. 5 in the main article as the values of $n_{\rm his}$ that simultaneously satisfy eq. (S29) and eq. (17) in the main article. For a given value of η_0 ($\equiv \lambda\rho_0\zeta/(k_{\rm on}^{\rm his}c_0\xi)$), there are two values of $n_{\rm his}$ that satisfy the spinodal condition, see the points labeled by sp1 and sp2 in fig. S1. We derive the critical point by finding the values of η_0, with which the two values of $n_{\rm his}$ on the spinodal curve are equal. The occupancy $n_{\rm his}$ thus satisfies eq. (S29), eq. (17) in the main article, and

$$\frac{d^2}{dn_{\rm his}^2}g(\eta)=0 \tag{S30}$$

at the critical point.

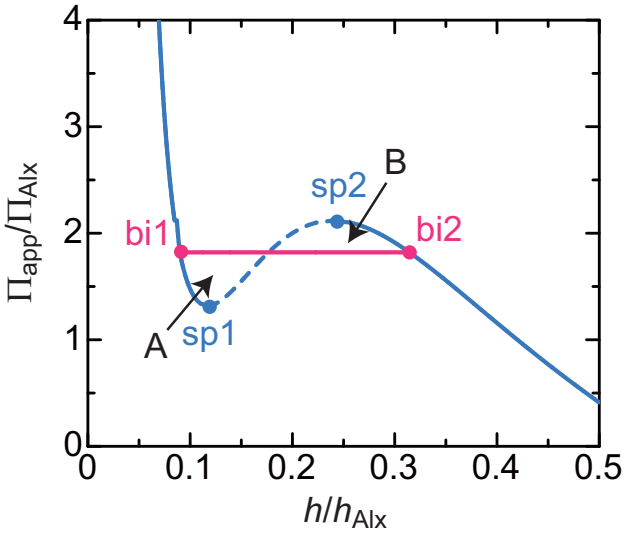

Figure S1: We use fig. 3**b** in the main article to derive the spinodal and binodal curves of fig. 5 in the main article (the cyan curve in fig. 3**b** is reproduced here). The spinodal curves are derived by the values of $n_{\rm his}$ that satisfy both eq. (S29) and eq. (17) in the main text (see the points labeled by sp1 and sp2). The magenta curve divide the Π-h curve so that the areas of the region A and B are equal. The latter condition ensures that the work that is necessary to gradually change the system between the two states (labeled by bi1 and bi2) that are at the two ends of the magenta line. The validity of this approach is discussed in sec. 4 in the main article.

S4 Coil-globule transitions

DNA brushes may show the coil-globule transitions for the cases that the force balance equation (eq. (15) in the main article or eq. (S18) in this ESI) has multiple solutions. When the combination n_- of the 2nd virial coefficients (defined below eq. (15)) is approximately unity, the right hand side (RHS) of eq. (15) in the main article is a monotonically increasing function of the brush height h for any values of the nucleosomal occupancy $n_{\rm his}$, see fig. S2 **a**. In contrast, when the combination n_- is smaller than a critical value (different from $n_-^{\rm c}$ that is defined in the main article), the RHS of eq. (15) in the main article is a non-monotonic function for large values of the nucleosomal occupancy $n_{\rm his}$, see fig. S2 **b**. The critical value

of the combination n_- (for the coil-globule transitions) has the form

$$n_-^* = 1 - \frac{2\tilde{u}}{(n_+ - 1)(1-\gamma)^{1/2}}, \tag{S31}$$

where $\tilde{u}$ is defined by $4u\sigma N_0/(3wh_{\rm Alx})$. Eq. (S31) is derived from the condition that the minimum of the first derivative of eq. (15) (in the main article) with respect to h becomes zero for the cases that the occupancy $n_{\rm his}$ is unity. The specific set of parameters ($n_+ = 1.8$, $\gamma = 0.7$, and $4u\sigma N_0/(3wh_{\rm Alx}) = 2.0 \times 10^{-3}$) that are used in the main article, $n_-^* \simeq 0.957$; the coil-globule transitions are not relevant for the values of the combination $n_- > 0.975$ that are used in the main article.

When pressures are applied to DNA brushes $\Pi_{\rm app} \geq 0$, the left hand side (LHS) of eq. (15) in the main article is negative; unless the local minimum of the RHS of this equation (see for example the black arrow in fig. S2 **b**) is negative, the coil-globule transitions are not driven by applied pressures. For relatively large brush heights, the first and second terms of eq. (15) in the main article dominate the third term of the equation. In such cases, the local minimum (of the RHS of eq. (15) in the main article) has an approximate form

$$\frac{1}{2^{2/3}} \frac{(n_+ - n_{\rm his})^{1/3}(n_{\rm his} - n_-)^{1/3}}{(1-\gamma n_{\rm his})^{4/3}} \tag{S32}$$

at a brush height $h_{\rm min}/h_{\rm Alx}$ that has the form

$$\frac{h_{\rm min}}{h_{\rm Alx}} \simeq 2^{1/3}(n_+ - n_{\rm his})^{1/3}(n_{\rm his} - n_-)^{1/3}(1-\gamma n_{\rm his})^{2/3}. \tag{S33}$$

This approximation is effective for

$$\frac{3}{2^{4/3}} \frac{\tilde{u}}{(n_+ - n_{\rm his})^{4/3}(n_{\rm his} - n_-)^{4/3}(1-\gamma n_{\rm his})^{2/3}} < 1; \tag{S34}$$

the RHS of eq. (15) in the main article is thus positive unless $n_{\rm his} \sim n_-$.

The asymptotic limit $n_{\rm his} \sim n_-$ corresponds to the cases that the brush height and the nucleosomal occupancy is close to the critical condition for the coil-globule transitions; the first and second derivatives of the RHS of eq. (15) (in the main article) with respect to the brush height h are zero. The latter condition leads to the critical nucleosomal occupancy, $n_{\rm his}^{\rm cg,c} = n_- + \delta n_{\rm cg,c}$, and the critical brush height $h_{\rm cg,c}$, where $\delta n_{\rm cg,c}$ and $h_{\rm cg,c}$ have the approximate forms

$$\delta n_{\rm cg,c} \simeq \frac{2\tilde{u}^{3/4}}{(n_+ - n_-)(1-\gamma n_-)^{1/2}} \tag{S35}$$

$$\frac{h_{\rm cg,c}}{h_{\rm Alx}} \simeq \tilde{u}^{1/4}(1-\gamma n_-)^{1/2}. \tag{S36}$$

We used an approximation $\delta n_{\rm cg,c}/n_- < 1$ to derive eqs. (S35) and (S36). With eqs. (S35) and (S36), the RHS of eq. (15) in the main article for the critical condition has the form

$$\frac{2\tilde{u}^{1/4}}{(1-\gamma n_-)^{3/2}}, \tag{S37}$$

which is positive (and may be small for $\tilde{u} \ll 1$).

The RHS of eq. (15) in the main article has a local minimum when the nucleosomal occupancy $n_{\rm his}$ is larger than the critical value $n_{\rm his}^{\rm cg,c}$. For the cases that the nucleosomal occupancy $n_{\rm his} = n_- + \delta n_{\rm his}$ is still not much larger than the combination n_- ($\delta n_{\rm his}/n_- < 1$), the brush height at the local minimum reads $h = h_{\rm cg,c} + \delta h$, where δh has an approximate form

$$\delta h \simeq \sqrt{\frac{2}{3}} \frac{(\delta n_{\rm his}^4 - \delta n_{\rm cg,c}^4)^{1/2}}{(n_+ - n_-)\delta n_{\rm his}^3}. \tag{S38}$$

With this approximation, the local minimum of the RHS of eq. (15) (in the main article) has the form

$$\frac{(n_+ - n_-)^3}{8\tilde{u}^2} \frac{\delta n_{\rm his}^4 + \delta n_{\rm cg,c}^4}{\delta n_{\rm his}} \left[1 - \frac{1}{\sqrt{6}} \frac{(\delta n_{\rm his}^4 - \delta n_{\rm cg,c}^4)^{3/2}}{\delta n_{\rm his}^4 + \delta n_{\rm cg,c}^4} \frac{1}{\delta n_{\rm his}^2} \right], \tag{S39}$$

which is positive because $\delta n_{\mathrm{cg,c}} \leq \delta n_{\mathrm{cg,c}}/n_- < 1$. Eqs. (S33) and (S39) suggest that the local minimum of the RHS of eq. (15) in the main article is always positive and thus the coil-globule transitions are not relevant (unless tensions are somehow applied to the DNA chains of brushes).

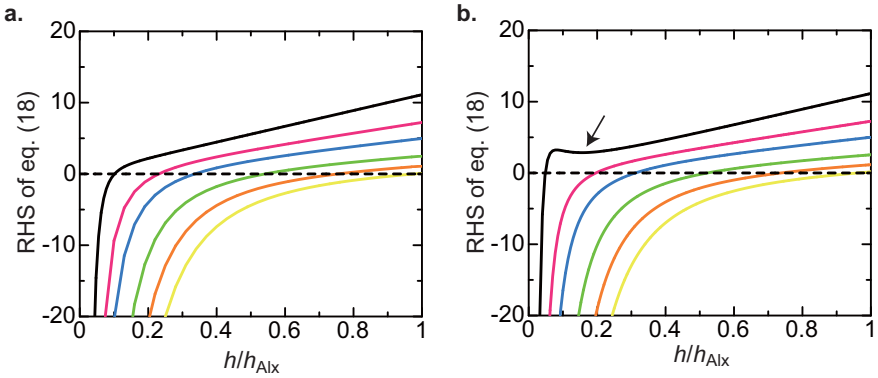

Figure S2: The right hand side (RHS) of eq. (15) in the main article is shown as a function of the rescaled brush height h/h_{Alx} for $n_{\mathrm{his}} = 0.2$ (yellow), 0.4 (orange), 0.6 (light green), 0.8 (cyan), 0.9 (magenta), and 1.0 (black). The combination n_- of the 2nd virial coefficients (defined below eq. (15) in the main article) is 0.99 (**a**) and 0.95 (**b**). The other parameters are fixed to $n_+ = 1.8$, $\gamma = 0.7$, and $4u\sigma N_0/(3wh_{\mathrm{Alx}}) = 2.0 \times 10^{-3}$. h_{Alx} $(\equiv N_0 l_{\mathrm{a}}(w\sigma/(6l_{\mathrm{a}}))^{1/3})$ is the length scale of the brush height.

References

(1) Yamamoto, T.; Safran, S. A. Transcription rates in DNA brushes. *Soft Matter* **2015,** 11, 3017-3021.