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Supporting Material

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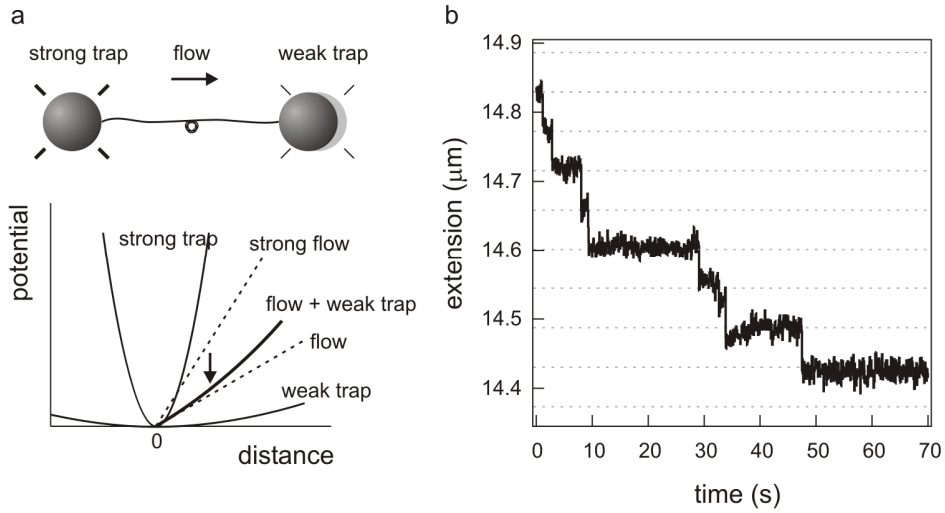
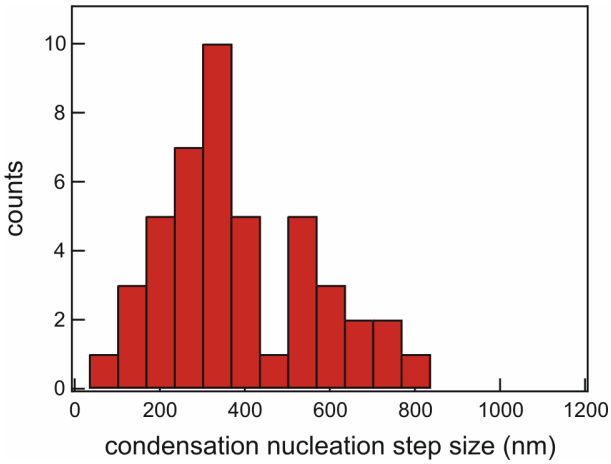


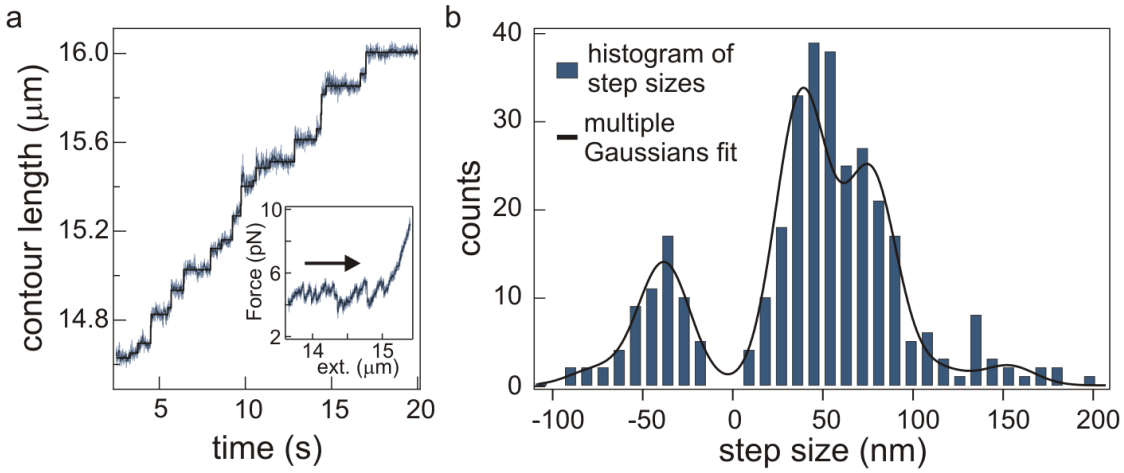
Supporting Figure S1: DNA condensation against a nearly constant force. (a) When a DNA condensing experiment as described in the main text is performed under constant force conditions (one bead in an optical trap, one bead in flow), a substantial fraction (>>50%) of the DNA is abruptly (< 500 ms) condensed (Supporting Movie M2). This is consistent with the nucleation-limited condensation as found in earlier constant-force experiments (1, 2). In order to slow down DNA condensation against a constant force, a slightly parabolically rising potential was created by applying a second optical trap with substantially reduced stiffness (~0.3 pN/μm). The maximum applicable force with such a weak trap is too low to oppose the pulling force of the DNA condensate. Therefore, the required force was predominantly created by added hydrodynamic flow on the free bead. The superimposed weak trap causes a slight increase in tension with distance from the trap center. **(b)** When the drag force on the DNA is reduced below a threshold value, stepwise condensation is observed. As the decrease in DNA extension pulls the bead out of the trap center, the force increases just enough to put up a small energy barrier preventing the wrapping of the next loop. This way, consecutive condensation steps, with a mean step size of ~50 nm, were observed until the bead was completely pulled out of the optical trap. Further condensation proceeded at the usual rapid pace (not shown).

1. Besteman, K., S. Hage, N. H. Dekker, and S. G. Lemay. 2007. Role of Tension and Twist in Single-Molecule DNA Condensation. Phys Rev Lett 98:058103.

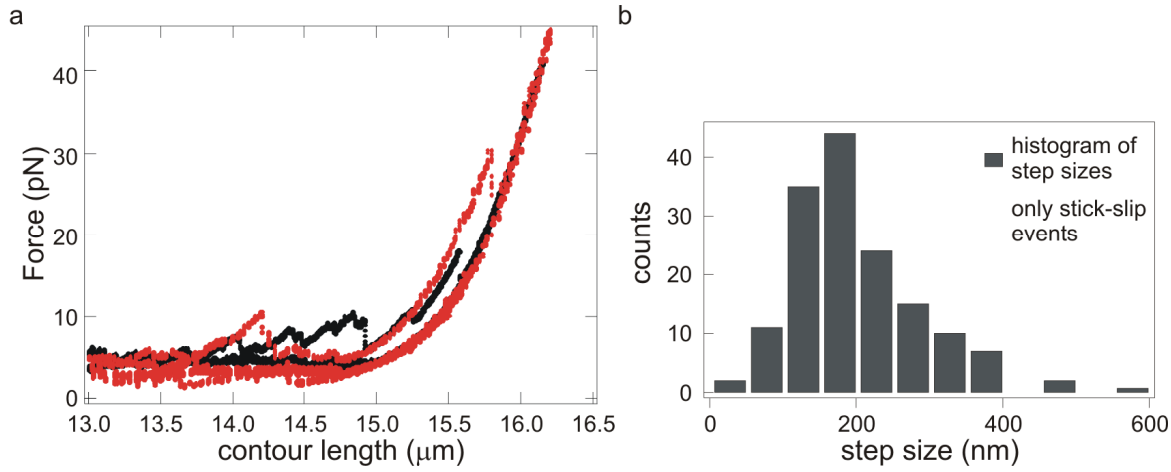
2. Su, T. J., E. Theofanidou, J. Arlt, D. T. Dryden, and J. Crain. 2004. Single molecule fluorescence imaging and its application to the study of DNA condensation. J Fluoresc 14:65-69.



Supporting Figure S2: Distribution of step sizes of condensation nucleation. Displayed is the contour length of DNA incorporated in the first condensate formed upon relaxing DNA under tension (registered by a force jump, step *B* in Fig. 2 of the main text). The data in the histogram is taken from force-extension traces with a DNA relaxation rate slower than 250 nm/s.



Supporting Figure S3: Stepwise disruption of DNA toroids. (a) Contour length plot of forced stepwise de-condensing of DNA. Like the condensation process, DNA de-condensation proceeds in discrete steps. The solid line shows the output of the step fitting algorithm. The inset displays the force-extension curve corresponding to the data. (**b)** Histograms of disruption step sizes, acquired from multiple de-condensation traces on several DNA molecules. Steps were analyzed by plateau fitting method. The distribution is quantitatively very similar to the toroid formation histograms in Figure 4b. Fitting with the same function as used for stepwise condensation results in $x_0 = 39 \pm 1$ nm, $\sigma = 20$ nm (normalized $\chi^2 = 1.2$), virtually the same parameters as found for condensing DNA.



Supporting Figure S4: Stick-slip events. (a) High-resolution force-extension curves of λ-DNA in the presence of >1 mM spermine that exhibit modest stick-slip behavior. **(b)** Distribution of stick-release events, analyzed by plateau fitting. Notice the difference in most frequent step size with that of Fig. 4b in the main text. The stepwise unraveling of condensed DNA in the case of high concentrations is apparently dominated by the enhanced attraction between DNA tracts in the toroid and causes disruption to occur in irregular large steps comprising multiple toroid loops.