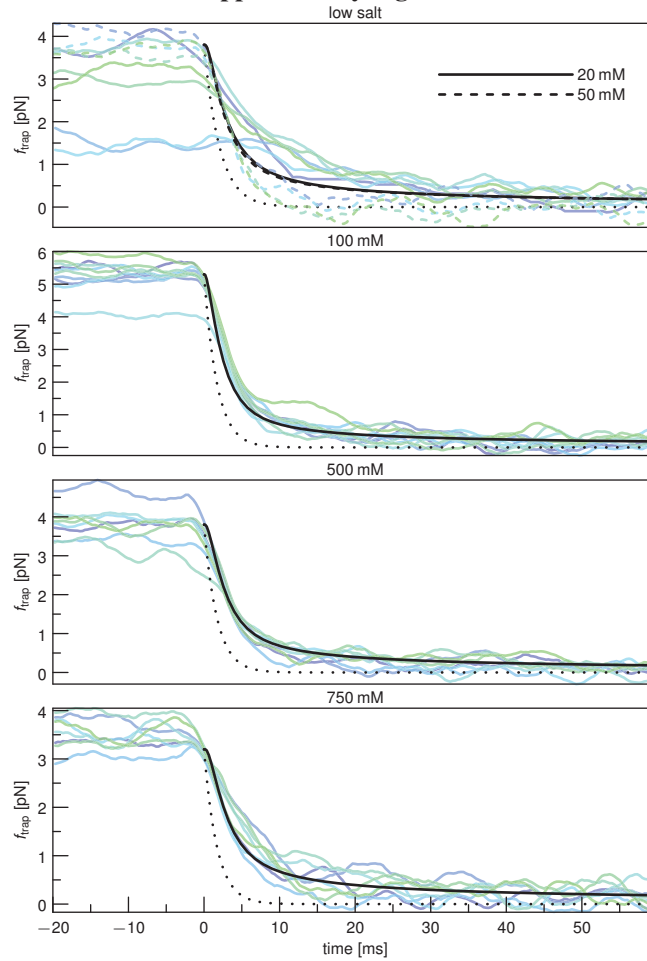


Rapid internal contraction boosts DNA friction

Supplementary Information

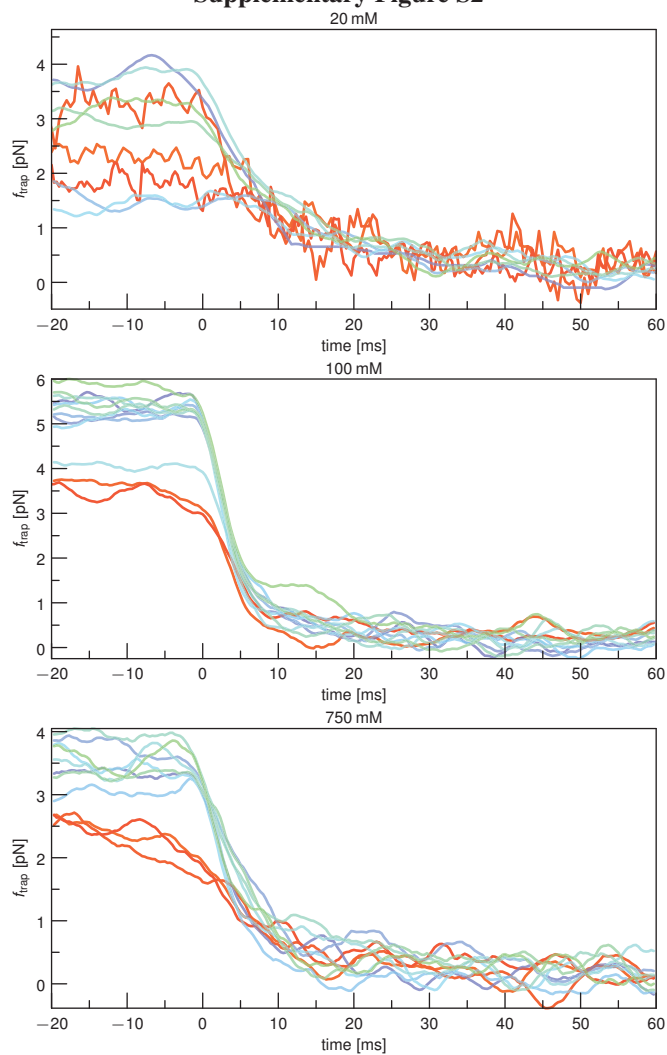
Oliver Otto, Sebastian Sturm, Nandanai Laohakunakorn,
Ulrich Keyser and Klaus Kroy

Supplementary Figure S1



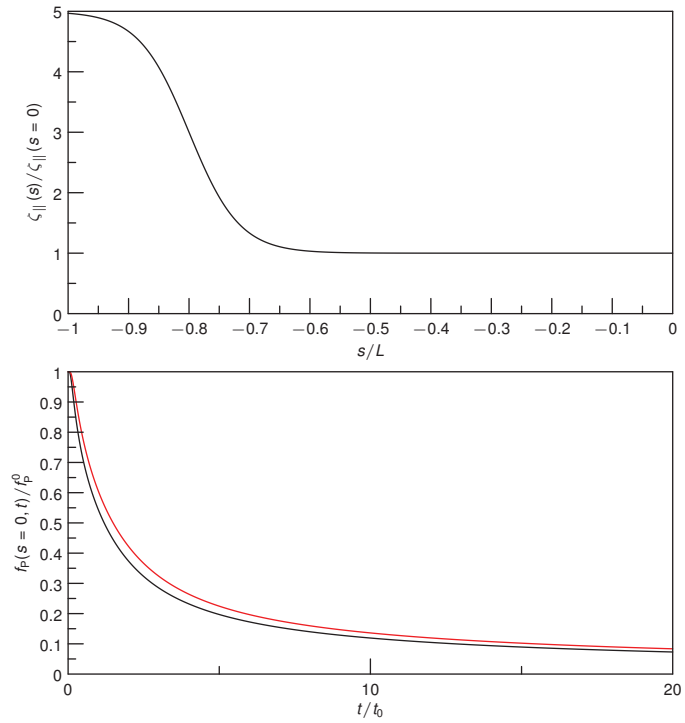
The time-dependent force $f_{\text{trap}}(t)$ on the bead. The “true” persistence length of DNA and its dependence on salt concentration is a somewhat disputed subject. In fig. 4, we have relied on the analytical approximation provided by Manning [33]; however, this assumption is not critical to our results as the above theoretical predictions for a constant persistence length of $\ell_p = 50$ nm show.

Supplementary Figure S2



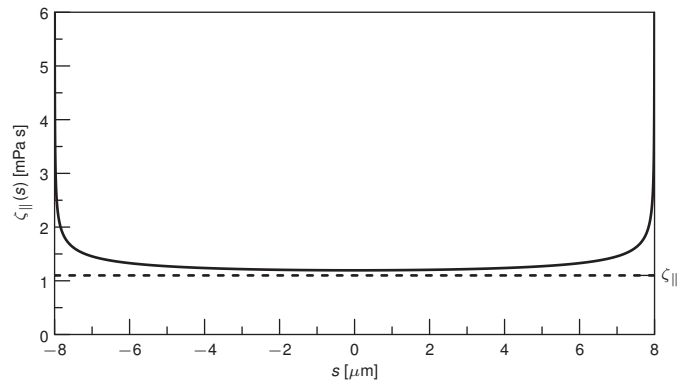
Control measurement. Instead of switching off the electric stretching potential, relaxation can also be induced by slowly (pulling velocity = $0.5 \mu\text{m s}^{-1}$) pulling the stretched polymer out of the nanocapillary. Red curves show the corresponding relaxation curves, as compared to our original data (blue curves, taken from fig. 4 in the main text).

Supplementary Figure S3



Additional hydrodynamic friction caused by the pore walls. To arrive at an upper bound to the additional frictional force brought about by hydrodynamic polymer-pore interactions, we have increased ζ_{\parallel} by a factor of 5 along the leftmost 20% of contour length (upper panel); even though this severely overestimates the effect, its influence on the resulting relaxation curve remains small (lower panel). Lower panel, black curve: original force relaxation curve. Red curve: force relaxation curve pertaining to the arclength-dependent friction coefficient shown above.

Supplementary Figure S4



Additional hydrodynamic friction caused by boundary effects. Since the Batchelor approximation pertains to an “infinitely long” cylinder, any real object of finite length will experience frictional forces that are (slightly) larger than the Batchelor result. To arrive at an upper bound to this additional retardation, we have used FEM simulations to compute the friction coefficient per length for a cylinder of diameter $d = 100\text{ nm}$ (solid line). Dashed line: corresponding Batchelor approximation $\zeta_{||}$. Though our computation overestimates the molecular diameter, and thus the magnitude of boundary contributions, the resulting additional friction is much weaker than the fictional pore-wall friction assumed in fig. S3.

Supplementary Methods

The following code iterates in v_{bead} to obtain the time- and space-dependent tension profile $f(s,t)$ (requires Wolfram *Mathematica*)

```
Needs["DifferentialEquations`InterpolatingFunctionAnatomy"];
Clear[fSol, maxT];
fSol[chi_, alpha_, tmax_] := fSol[chi, alpha, tmax] =
Module[{solFromG, gFromF, eqnSys, iterateG, g, iterations = 1, maxDeviation = Infinity, newSol, oldSol, times,
startingGuess, temp, cachedValues, closestMatch, closestF, initialProfile, maxIterations = 500, deviationLimit,
superpositionFactor, maxF, firstsol, ndopts},
ndopts = {MaxSteps -> 10000};
superpositionFactor = 0.5;
deviationLimit = 1 *^-3;
g[0] = -6 #/(1 + 9#^2)^(4/3) &;
eqnSys[g_, f_, s_, t_] := {f[-1, t] == 0,
Derivative[1, 0][f][0, t] == g[t],
Derivative[2, 0][f][s, t] == Derivative[0, 1][f][s, t] / f[s, t]^(3/2),
f[s, 0] == 1 - Exp[-(1 + s)*50] (* approximate initial tension profile, details irrelevant *)};
solFromG[g_] := Module[{s, t, f, sol, z},
sol = Flatten@NDSolve[eqnSys[g, f, s, t], {f}, {s, -1, 0}, {t, 0, tmax}, Sequence@@ndopts];
{Function@@{{t}, Derivative[1, 0][f][0, t] /. sol}, Function@@{{t}, f[0, t] /. sol}}];
iterateG[g_] := Block[{t}, Function@@{{t}, (superpositionFactor gFromF[solFromG[g][[2]]][t] + (1 -
superpositionFactor)g[t])}];

g[n_Integer] := g[n] = iterateG[g[n - 1]];
gFromF[f_] := Block[{t, G, sol},
sol = Flatten@NDSolve[{G[0] == 0, G'[t] == -alpha f'[t] - chi G[t]}, G, {t, 0, tmax}, Sequence@@ndopts];
G /. sol];

times = Table[t, {t, 0, tmax, tmax/400}];
oldSol = solFromG[g[0]][[2]];
While[iterations < 3 || (maxDeviation > deviationLimit && iterations < maxIterations),

newSol = Check[solFromG[g[iterations++]][[2]], $Failed, {NDSolve::mxst}];
maxDeviation = If[newSol === $Failed, Infinity, Max[Abs[Re[newSol[#]] - oldSol[#]]] & /@ times];
maxF = Max[Re[newSol[#]]] & /@ times];
If [maxDeviation > 1 || maxF > 1.1,
(* failure : take one step back, try slower approach *)
g[( iterations --) - 1] =.;
superpositionFactor = 0.5 * superpositionFactor;
If [superpositionFactor <= 1 *^-10, iterations = maxIterations],
oldSol = newSol];
If [ iterations < maxIterations, newSol, $Failed];
fSol[chi_?NumericQ, alpha_?NumericQ] := fSol[chi, alpha] = TimeConstrained[Check[Module[{sol = None, tmax
= 10, s, t},
While[(sol === None || sol[tmax] > 0.1) && !(sol === $Failed),
tmax = tmax * If[NumericQ[sol[tmax]], Max[2, Ceiling[1.5/(1 - sol[tmax])]]], 2];
sol = fSol[chi, alpha, tmax];
maxT[chi, alpha] = tmax;
If [sol === $Failed, $Failed,
```

```
Function@@{{t}, Piecewise[{{sol[t], t <= tmax }, 0]}], $Failed, 1000, $Failed];
```

Supplementary Note 1: Hydrodynamic corrections

At the outset of our experiment, several micrometers of the strand of DNA are still located within the capillary and should thus experience greater hydrodynamic friction, surpassing the drag coefficient in free solution by a factor $\sim \log(L/d)/\log(W/d)$, where W denotes the distance between polymer and capillary wall. With a minimum wall distance on the order of only ~ 20 nm we may see a relative increase in ζ_{\parallel} on the order of 3 – 5. To roughly estimate the effect this might have on the measured tension $f(s=L, t)$, we replaced ζ_{\parallel} by a fictional friction profile $\zeta_{\parallel}(s)$ that coincides with the usual friction coefficient along the rightmost 80% of the polymer and is about fivefold larger along the remaining 20%. Obviously this overestimates the resultant changes, since in reality the polymer does not constantly feel an elevated frictional resistance at its free end, but instead escapes from the region of increased drag, thus progressively returning to the original, lower, friction coefficient. Still, we see no more than a 15% increase in the effective relaxation time, see Supplementary Figure S3. To make sure that no significant retardation is caused by the capillary entrance, we performed a second set of relaxation experiments at full stretching voltage, instead triggering relaxation by slowly pulling out the stretched DNA using the Piezo stage, see Supplementary Figure S2.

Apart from polymer-wall interactions, the friction coefficient ζ_{\parallel} is in itself only a lowest-order approximation to the true hydrodynamics involved. In reality, the hydrodynamic drag force per length should attain its infinite-length limit

$$\zeta_{\parallel} = \frac{2\pi\eta}{\log(L/d)}$$

only within the polymer bulk and increase towards the ends, as outlying parts of the polymer receive less help from their surroundings in setting the solvent in motion. However, as Supplementary Figure S4 shows, this increase is limited to a narrow boundary layer (where we have chosen a larger molecular diameter $d = 100$ nm to account for more moderate aspect ratios than realized in our setup; in our case, the molecular diameter measures only $d \approx 2$ nm, thus rendering the boundary effect even weaker than indicated by fig. S4) and thus probably no more troubling than the neglect of flow gradients or the coarse-graining of molecular conformations inherent in the derivation of ζ_{\parallel} . We therefore take the view that the constant-drag coefficient approximation cannot significantly be improved upon without resorting to full-scale simulations.

Supplementary Note 2: Stem-flower effect

Given the conical shape of the nanocapillary (save for a very short cylindrical bottleneck at the entrance), by eq. (3) in the main text we have $f_0(s) \propto s^3$ along the first $3\ \mu\text{m}$ of DNA. Under a typical prestretching force of about $4\ \text{pN}$, the critical force threshold of $0.08\ \text{pN}$ required to pull the polymer contour taut is thus reached a distance $\ell \approx 3\ \mu\text{m}(0.1/4)^{1/3} \approx 900\ \text{nm}$ from the free polymer end. Within this outermost micrometer, the weakly-bending approximation breaks down, instead giving way to an almost force-free coil or “flower”. As soon as the potential is switched off, however, this flower vanishes as tension within the polymer is now generated by hydrodynamic drag forces. In contrast to the initially applied electric field, the latter increase towards the free end. Given the roughly parabolic profile of $f(s,t)$, the size of the “tension-free” region close to the retracting end measures only roughly $200\ \text{nm}$, or two Kuhn lengths, at the outset and increases to about 600 to $700\ \text{nm}$ until the terminal tension has dropped to $1\ \text{pN}$. Even this larger value corresponds to a transverse fluctuation amplitude on the order of $\sqrt{Nb} \approx \sqrt{7} \cdot 100\ \text{nm} \approx 250\ \text{nm}$, locally increasing ζ_{\parallel} by a factor $\sim 2 - 3$, a much weaker effect than the (putative) hydrodynamic polymer-wall interactions investigated above.