# Stretching of surface-tethered polymers in pressure-driven flow under confinement

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## 1 Simulation details

We used a mesoscopic simulation approach by combining a coarse-grained polymer model with a lattice-Boltzmann fluid. The coarse-grained MD simulations were performed with ESPResSo using a simple bead-spring model. Inter-bead interactions were modeled with Weeks-Chandler-Andersen potential and adjacent beads were connected by a finitely extensible nonlinear elastic bond potential. Although not containing charges, this model resembles a polyelectrolyte in aquous solution containing salt (e.g. double-stranded DNA molecules in buffer solution with high concentration of salt) reasonably well. Single beads in coarse-grained models usually represent the individual persistence lengths within a polymer<sup>1</sup>. For  $\lambda$ -DNA, a persistence length of  $(42 \pm 5)$ nm was found<sup>2</sup>, resulting in a bead diameter  $b \approx 42$  nm. Electrostatic interactions in aqueous solution usually decay on a scale between  $\lambda_B = 0.7$ nm, where  $\lambda_B$  is the Bjerrum length<sup>3</sup>, and several nanometers for low concentrated salt solutions. The electrostatic screening length can be expressed in terms of the Bjerrum length  $\lambda_B$ 

$$\lambda_D = \frac{1}{\sqrt{4\pi\lambda_B\sum_i c_i z_i^2}},\tag{1}$$

with  $c_i$ ,  $z_i$  being the concentration and valency of charged species *i*. Eq. (1) shows that the length scale on which electrostatic interactions decay are much smaller than the persistence length for the experimentally employed parameter set such that electrostatic interactions can be neglected if we are only interested in the scaling behaviour of the polymer we want to model.

Hydrodynamic interactions were included by coupling the beads to a thermalized lattice-Boltzmann (LB) fluid. The grid spacing of the LB fluid in a D3Q19 arrangement is a = b. The coupling between the LB fluid and the MD beads is achieved by the standard scheme<sup>4</sup> according to

$$\vec{F}_c(\vec{r}) = \Gamma(\vec{v}_m(\vec{r}) - \vec{u}(\vec{r})), \qquad (2)$$

where  $\vec{F}_c$  is the coupling force on a bead,  $\Gamma = 20m_0/t$  the bare coupling constant, and  $\vec{v}_m$  and  $\vec{u}$  are the velocities of the beads

and the fluid, respectively. The fluid velocity is calculated by a linear interpolation on the eight nearest neighbors. An equal and opposite force is applied to the fluid using the same weights used for the linear interpolation of the fluid velocity. For the lattice-Boltzmann simulations, the fluid and the coupling with the MD beads are thermalized using a lattice-Boltzmann thermostat such that  $T = \varepsilon = k_B T$ , where  $\varepsilon$  is the energetic prefactor. The time step for both the lattice-Boltzmann fluid and the MD integration is set to  $\Delta t = 0.01t$ , where *t* is the time scale used for the fluid properties. Two infinitely long plates at  $\pm z_B$  with varying plate distances *H* represent the physical boundaries of the microchannel. The plates inter-



**Fig. 1** Schematic drawing of a polymer (black spheres) tethered to a channel wall (gray area) under the influence of a planar Poisuille flow (blue grid).

act via the same WCA potential as described above with the polymer beads and via bounce-back interactions to implement no-slip boundaries with the fluid<sup>5</sup>. We fixed the first bead at  $-z_B$  in order to model a tethered polymer. The equivalent of a pressure gradient along the channel is accomplished by an external force density according to  $\partial p/\partial x = -\rho f_x/m_0$  applied onto the LB fluid nodes in *x*-direction, resulting in a parabolic Poiseuille velocity profile. We study the influence of external forces in the range between 0.01 and 1.0  $\frac{1}{bt^2}$ . A schematic illustration of our simulation setup is shown in figure 1.

# 2 Scaling of the fractional extension in the strongly stretched conformation

According to the scaling argument presented by Ladoux and Doyle<sup>6</sup>, the fractional extension of a surface-tethered polymer chain, exposed to a linear shear flow, is given by  $1 - \frac{x}{L} \sim \dot{\gamma}_{wall}^{-\frac{1}{3}}$ 

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in the strong stretching limit, where x is the extension of the chain, L is the contour length and  $\dot{\gamma}_{wall}$  is the wall shear rate. If the viscosity of the buffer is constant, then  $\dot{\gamma}_{wall}$  can be replaced by the wall shear stress  $\tau_{wall}$ . Figure 2a shows the experimentally determined scaling of the fractional extension of  $\lambda$ -DNA molecules with the applied wall shear stress, assuming the contour length  $L = 20.4 \mu$ m. This value of the contour length is determined from the staining ratio<sup>7</sup> (1 dye molecule per 5 base pairs) used in our experiment. The figure indicates that the fractional extension of the tethered DNA molecules in the strong stretching limit can be expressed by the Ladoux and Doyle scaling. After having verified the validity of this scaling in the strong stretching limit at different degrees of confinement, we employed the scaling relationship to estimate the contour length more accurately. The variation of the fractional extension with the wall shear stress, based on the values of the contour length determined that way, is shown in figure 2b.

# 3 Scaling of the fractional extension of $\lambda$ -DNA for small values of the wall shear stress

The fractional extension of surface-tetehered  $\lambda$ -DNA molecules at small values of the wall shear stress is expressed by the relation  $\frac{x}{L} \sim \tau_{wall}^{m}$ , where *m* is a scaling exponent. We determined the value of *m* corresponding to different channel heights by fitting the fractional extension data at small  $\tau$  in a double-logarithmic plot with a straight line. The resulting values of of *m* are indicated in the legend of figure 3.

#### 4 Angle of orientation of $\lambda$ -DNA

In figure 4 the angle of orientation of surface-tethered  $\lambda$ -DNA at different channel heights is plotted over the entire range of the wall shear stress applied in our experiments. We also include the plot of the angle of orientation in an effectively unbounded situation, as described in the work of Lueth and Shaqfeh<sup>8</sup>, to compare the variation of the orientation angle under confinement to that in an unconfined situation.

#### **5** Quantification of the data collapse

Let  $\Delta e$  be the width of the error bar (indicating the standard deviation) in fractional extension at a specific value of  $\tau_{wall}L$ . At the same value of  $\tau_{wall}L$ ,  $\Delta v$  is the standard deviation of the data set of fractional extension values corresponding to chains of different contour length. We use the ratio  $\frac{\Delta v}{\Delta e}$  as an indicator for the collapsing of the different curves on a single master curve. If  $\frac{\Delta v}{\Delta e} \rightarrow 0$ , the collapse is perfect.

The distribution of  $\frac{\Delta v}{\Delta e}$  over the entire range of  $\tau_{wall}L$  is shown in the inset of figure 6a and b. The analysis shown





**Fig. 2** (a) Scaling of  $(1 - \frac{x}{L})$  with  $\tau_{wall}$  for  $\lambda$ -DNA assuming a constant contour length of 20.4 $\mu$ m. The black straight line indicates a slope of  $-\frac{1}{3}$ . (b) Plot of  $(1 - \frac{x}{L})$  with  $\tau_{wall}$  for  $\lambda$ -DNA using the contour lengths determined from the scaling of Ladoux and Doyle<sup>6</sup>. The black straight line indicates a slope of  $-\frac{1}{3}$ .

in figure 6a corresponds to figure 2b of the main article. We note that in almost 40% of the cases,  $0 < \frac{\Delta v}{\Delta e} < 1$ , whereas in almost 50% of the cases,  $4 < \frac{\Delta v}{\Delta e} < 12$ . This type of bimodal distribution of the ratio  $\frac{\Delta v}{\Delta e}$  indicates that the curves in figure 2b of the main article are slightly different, which we identify as a weak effect of confinement.

The histogram in figure 6b shows the frequency of different



Fig. 3 Fractional extension of  $\lambda$ -DNA as a function of wall shear stress at different channel heights. The plot is shown in double-logarithmic style, where the  $\frac{x}{L} \sim \tau_{wall}^m$  scaling expected at small shear-stress values translates to a linear curve.



Fig. 4 Angle of orientation of the surface tethered  $\lambda$ -DNA molecules plotted against the wall shear stress in channels of different heights.

values of  $\frac{\Delta v}{\Delta e}$ , corresponding to figure 6 of the main article. From the histogram, we see that values with  $0 < \frac{\Delta v}{\Delta e} < 1$  have a frequency of about 50%, while values with  $0 < \frac{\Delta v}{\Delta e} < 2$  have a frequency of about 75%. This indicates that the different curves collapse on each other within the range defined by the experimental error bars.



**Fig. 5** Schematic depicting the method of analyzing the collapse of two curves on each other.

### 6 Post-processing of confocal microscopy images

Confocal microscopy images of surface-tetehred  $\lambda$ -DNA molecules were captured with a resolution of  $256 \times 256$  pixels (0.25  $\mu$ m/pixel) in the x - z plane and a resolution of 0.1  $\mu$ m in y-direction (see figure 7). We could not take images with a higher resolution because of the increased time required to scan a molecule. An increased scanning time increases the probability of photo-induced breakage of the molecules. The molecules were stretched in x-direction by application of a pressure-driven flow. A side-view of a molecule (i.e. a view parallel to the x - y plane) was rendered by averaging the pixel intensities from different planes (parallel to the x - y plane) over a slab of width  $\Delta z$  slightly larger than the characteristic width of the molecular fluctuations in z-direction. Processing of the stacked images was done using a MATLAB script. From the images of a fluctuating molecule, we can estimate the average or most-probable angle of orientation  $\alpha$  by the angle between the tethering plane and a vector connecting the tethering point and the farthest fluorescing point (as shown in the inset of figure 5 of the main article). After rendering the side-view images, these vectors were identified for each molecule in a MATLAB image processing window by manually locating the two points defining the corresponding vectors.

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TETHERING POINT X Z

DIRECTION OF FLOW

Fig. 7 Confocal microscopy image of a tethered  $\lambda$ -DNA molecule in isometric view.

**Fig. 6** Normalized histograms of the frequency of  $\frac{\Delta v}{\Delta e}$ . The insets show the distributions of  $\frac{\Delta v}{\Delta e}$  over the entire range of  $\tau_{wall}L$ . (a) Data corresponding to figure 2b of the main article. (b) Data corresponding to figure 6 of the main article.

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