Supplemental Information

Interaction prolongated DNA translocation through solid-state nanopores

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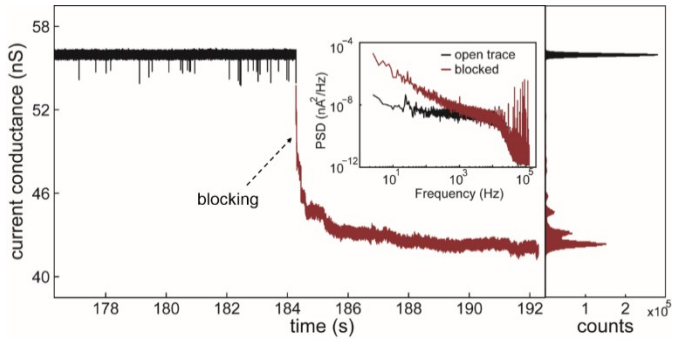

Figure S1. Raw current trace displaying the variation in fluctuation of baseline and noise level in a typical blocking incident. Standard events recording is shown in black while the blocked one in red. It can be intuitively told that after blocked, the fluctuation of current baseline obviously enhanced, which can be confirmed by the whole points histogram distribution of current baseline on the right. As a result, the clear baseline shows a well-fitted single peak component (black) while the blocked state occupied several small peaks with poor geometry. Inset is the power spectrum density (PSD) diagram calculated from these 8 second ionic current baselines indicates great increase in noise level among low frequency range (1~1000 Hz), which can be as much as 4 orders of magnitude. Meanwhile, along with the blocking process, no translocation events can be detected at all. To sum up, what happened in a typical blocking incident might be the analyte molecular clogging the pore entrance as a single mass of flexible bead or several polymers as a bunch, which cut off the travelling pathway for other molecules.

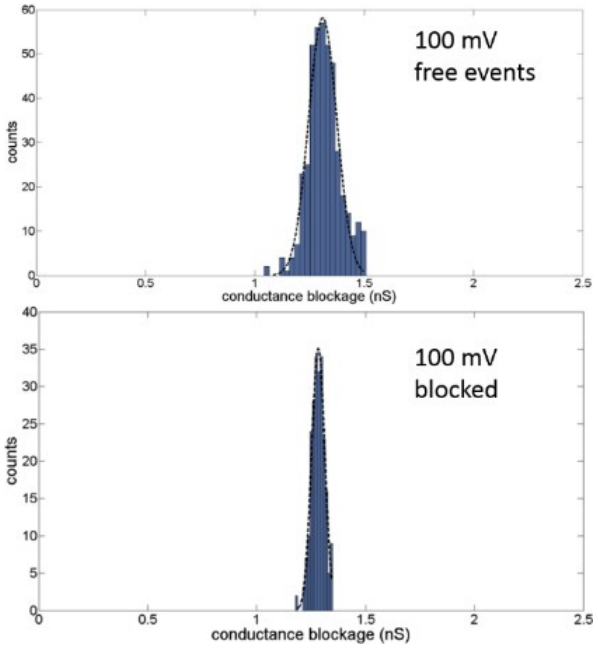

Figure S2. Conductance blockage Gaussian fit of 5 kbp dsDNA translocation under bias of 100 mV through open solid-state nanopore, and DNA-blocked pore as control, demonstrating the similar level of blockage, which suggests the captured events are caused by success DNA translocation.

length	**5 kbp**	**5 kbp**
voltage (mV)	100	100
free or blocked	free	blocked
number of events	422	332
blockage level of single (nS)	1.31±0.065	1.282±0.030
R-square	0.9519	0.9653
t_{peak} (μs)	132±4	328±67
exponential fit τ_{decay} (μs)	29±3	643±134
R-square	0.9412	0.9411
τ_{rel}	1±0.16	22.4±5.3
number of >2 ms	0	11

percentage of >2 ms	0	4.85 %
number of >1 ms	0	62
percentage of >1 ms	0	27.31 %

Table-S1. Detailed data processing and integration of all events distribution through a 8.8 nm nanopore for the data sets used in the main text.

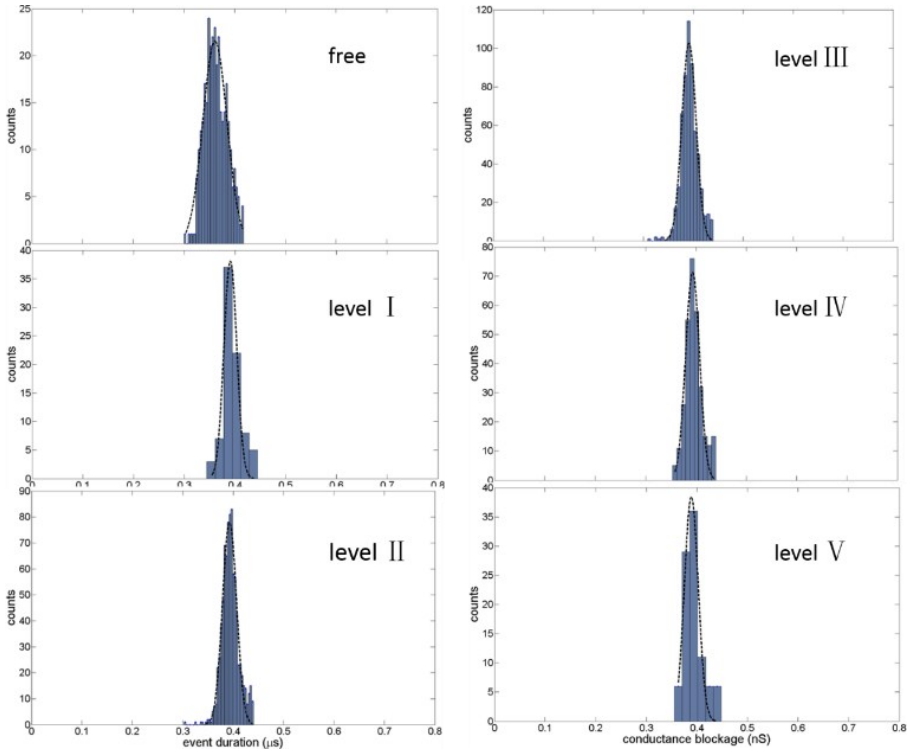

Figure S3. Conductance blockage Gaussian fit of 5 kbp dsDNA translocation under bias of 200 mV through open solid-state nanopore, and DNA-blocked pore at different current levels.

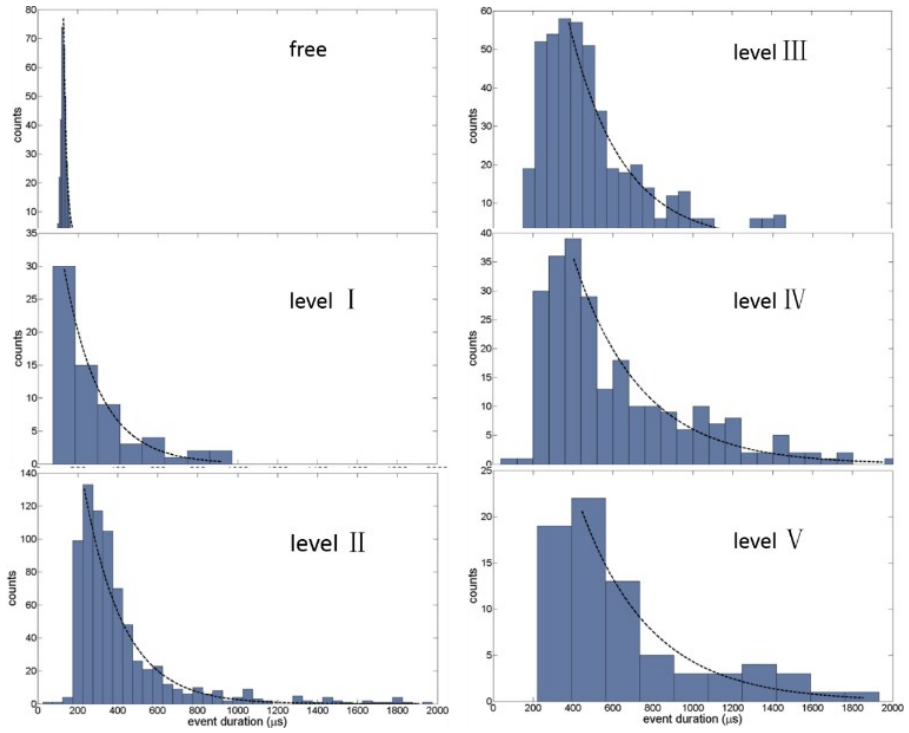

Figure S4. Event duration exponential fit of 5 kbp dsDNA translocation under bias of 200 mV through open solid-state nanopore, and DNA-blocked pore at different current levels.

S-5 Simulation of driving force by COMSOL

To evaluate the influence of the electroosmotic flow, we have calculated the driving force of the translocation DNA in different position when 0 and 3 DNA molecules is anchored respectively. The driving force is the resultant force of the electric field force and the electroosmotic flow.

The anchored DNA is positioned in the way shown in Figure S5a. All three anchored DNA is 0.2 nm away from the nanopore. And the translocation DNA is set at various position along the nanopore center. The result shows that the driving force increases when the translocation DNA is approaching to the nanopore wall (positive direction) or the anchored DNA (negative direction, as the arrow indicated in Figure S5b). It is mainly due to the no slip boundary condition that will decrease the electroosmotic flow and thus lead to a larger driving force.

Furthermore, the total driving force behaves tiny reduction when 3 DNA are anchored into the nanopore wall (blue curve in Figure S5b) compared to the situation of no anchored DNA (red curve in Figure S5b). For instance, in the case where the translocation DNA is 2 nm from the center, the driving force decreases from 22.84 pN to 21.36 pN, less than 6.5%, which is negligible compared with the 10 times extension in translocation duration.

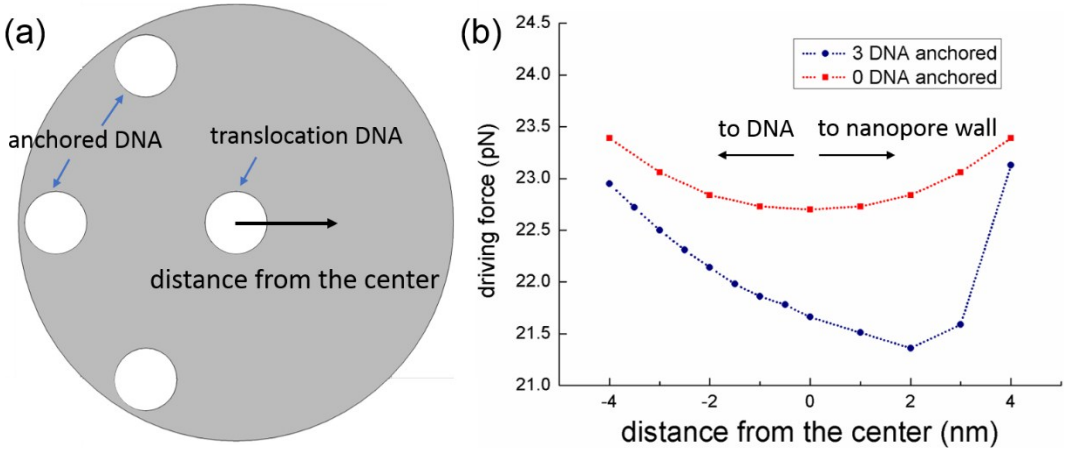

Figure S5. (a) Schematic diagram for driving force calculation, showing the cross section of a nanopore channel where 3 DNA are anchored (the hollow part is occupied by DNA), with the translocation DNA set at various position along the nanopore center. (b) Driving force as the resultant force of electric field force and electroosmotic flow as a function of the tranlocating DNA distance from nanopore center. 0 means the translocation DNA is at nanopore center, more positive position means the translocated DNA is more close to the nanopore surface (right), more negative position means the translocated DNA is more close to the anchored DNA surface (left), as the arrows indicate. Red squares and blue dots represent conditions with bare nanopore and pore anchored by 3 DNA, respectively.

S-6 Simulation of current blockage by COMSOL

As the time scale of DNA translocation process is much larger than that of ion relaxation toward equilibrium distribution, a Poisson-Boltzmann equation as well as a Nernst-Planck equation can describe the ionic concentration distribution. Then the

Poisson-Boltzmann equation is coupled with Navier-Stokes equations to calculate the fluid dynamics properties. Both the driving force exerted on translocated DNA and the ionic current can be numerically calculated in this model. To investigate the influence of multiple DNA polymers on the ionic current, we have calculated the value of ionic current with different number of DNA polymers inside the nanopore channel, ranging from 0 to 5. In simulations, we set that all of the DNA polymers inside are arranged in an axially symmetric way, and each polymer has a constant distance from the nanopore inner wall of 3 nm. For instance, for the situation of four DNA polymers inside, the cross section view of the nanopore channel is demonstrated as Figure S6a (The solid part is filled with solvent and demonstrates the geometry profile of the nanopore, and the hollow part is the space occupied by DNA polymers. Here a 1 nm radius cylinder is used to model the DNA polymer).

The ionic current simulation results show that the current drop between each current level is highly uniform as a function of the number of DNA polymers inside nanopore, showing a monotonic decrease as the number of anchored DNA increases, which agree very well with the experimental data (Figure S6b). Further simulations on the conductance blockage of DNA translocation on each level is identical on those five levels, which also fit very well with the experimental data (Figure S6c).

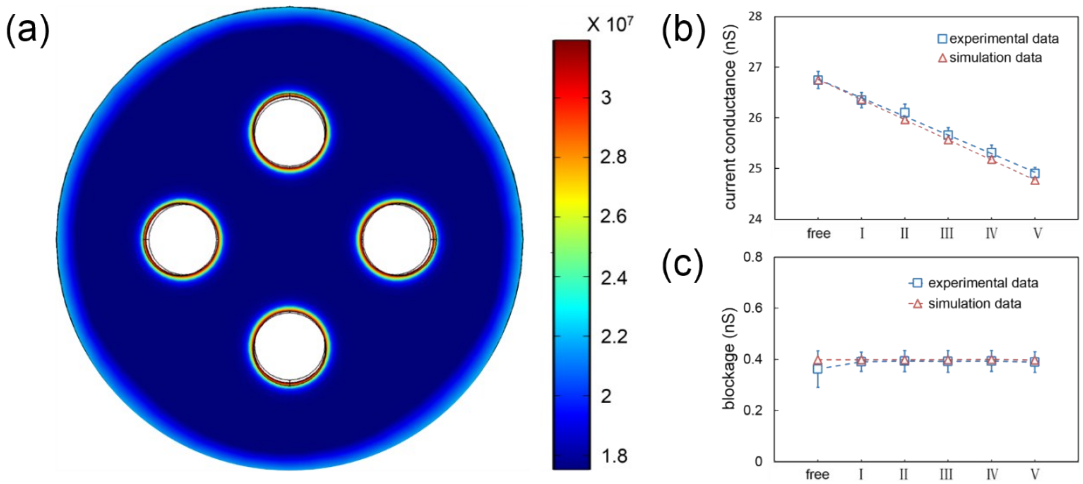

Figure S6. (a) The cross section of a nanopore with 4 DNA inside in Comsol simulation, color bar demonstrating the electrical field distribution in this system, unit is V/m. (b) The peak value of Gaussian fitted whole points histogram distribution is plotted as function of current level from free state to level V with blue square (error

bars: 95% confidence bounds), while the red triangles represent simulation results in a nanopore of identical size and geometry, with number of blocked DNA from 0 to 5. The dashed lines are linear fitted with R-square over 0.99. (c) Conductance blockage for translocation events of each level is graphed both in experimental data and simulation results. These two series have shown nice homogeneity in blockage level.

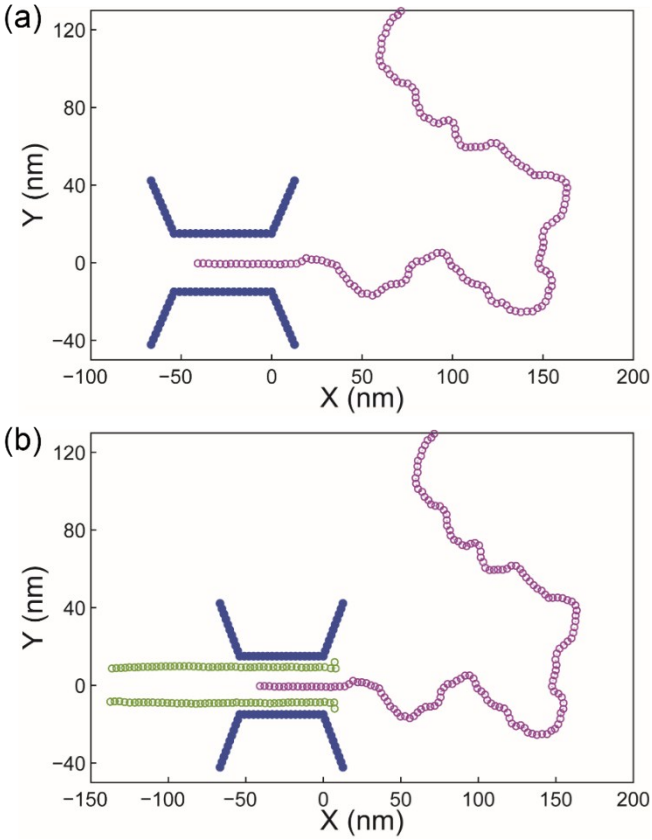

Figure S7. Schematic diagram for Langevin Dynamics simulation. (a) The geometry of the nanopore is set to be like an hourglass, according to the tomography image, with 150 nm membrane thickness, 50 nm effective length (length of central cylindrical region), and 7 nm pore radius. The green polymer chains in (b) are the anchored DNA whose right end is fixed outside the nanopore channel on the chip surface, and the violet polymer chain is the translocation DNA molecule.

Simulation considerations

Force and energy

1. Bond Stretching Interaction

We take a linear restoring force to model the stretching of covalent bonds on the DNA backbone. The bond energy is given by:

$$U_{bond} = k_{spring}(x-a)^2$$

The parameter k_{spring} depends on the drag coefficient along the tangent direction of the polymer contour γ_1 and the time step dt, which is given by:

$$k_{spring} = \frac{\gamma_1}{10 \cdot dt}$$

2. Excluded volume interaction

This interaction is employed to prevent the polymer chain from crossing and overlapping. The interaction is a pure repulsive force modeled by an interceptive linear force, and thus the potential energy is given by:

$$U_{excluded} = k_{spring}(r-a)^2 \qquad (r < a)$$

where r is the distance between any two beads, and the constant k_{spring} share the same value as it in the bond energy.

3. Chain Bend energy

Differ from the simulation of ssDNA, which has a very short persistence length and so a low stiffness, the dsDNA has a persistence length of 50 nm, which represents a very strong stiffness and rod-like property. This fact requires us to introduce a bending energy to confine the chain from bending. Considered the bend angle between 3 adjacent beads is very tiny, it is reasonable to apply a square potential given by:

$$U_{bend} = k_{bend}(\theta-\theta_0)^2$$

To determine the value of k_{bend}, we have run a series of simulation with different value of k_{bend} and inspect the stiffness of the polymer chain characterized by persistence length. The definition of persistence length P is:

$$\langle \cos\theta \rangle = e^{-(L/P)}$$

Where L is the distance between the bead i and bead j along the contour of the chain,

and θ is the angle between the tangent vector at bead i and bead j. The k_{bend} is adjusted to 8.05E-21 J in order to make the chain's persistence length accord with 50 nm, according to Table S2.

Table S2. The persistence length of dsDNA depending on k_{bend}.

k_{bend} (J)	4.60E-21	5.75E-21	6.90E-21	8.05E-21	9.20E-21
persistence length (nm)	21.6	35.1	41.4	49.1	55.7

4. DNA-nanopore Interaction

The interaction between DNA and nanopore in our model is purely repulsive, and the type and magnitude is same with the excluded volume interaction. It is given by:

$$U_{wall-DNA} = k_{spring}(x-a)^2 \qquad (x<a)$$

DNA initial configuration

In this model, due to the existence of an excluded volume interaction, a self-avoiding random walk is needed to create the initial configuration of DNA molecules. The first bead position is settled at the entrance of the nanopore channel on the cis side and then we start random walking towards the Cis side to create the initial configuration. After making a new step, we place a bead on that position and connected the new bead with the previous one with a spring. Each step covers a distance of *a*, and the direction of each step depends on the direction of the last step plus a random term. For instance, the step i is towards the direction of $\vec{e}_i$, so the step i+1 direct towards mainly at the $\vec{e}_i$ direction but with a fluctuation of the angle due to the random term which follows a $[-\theta,\theta]$ even distribution. To determine the value of θ, we create a series of 1000 chain configurations with different θ value, and adjust the θ until the configurations give a persistence length of 50 nm statistically.

After creating the equilibrium configurations outside the pore channel, we further add a few beads to the head of the polymer chain that filled in the pore channel. This is to prevent the polymer from escaping from the nanopore and causing unsuccessful

translocation events. In our model, the major purpose is to explore the dynamic process of translocation but not the capture process that DNA trapping into the pore. Therefore, we put the beads into the pore at the beginning of simulation and ignore the capture process which is very short compared to the translocation duration.