Supporting material to "Characterizing and Controlling the Motion of ssDNA in a Solid-State Nanopore"

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Ratcheting ssDNA in the DNA transistor solvated with a 1 M NaCl electrolyte

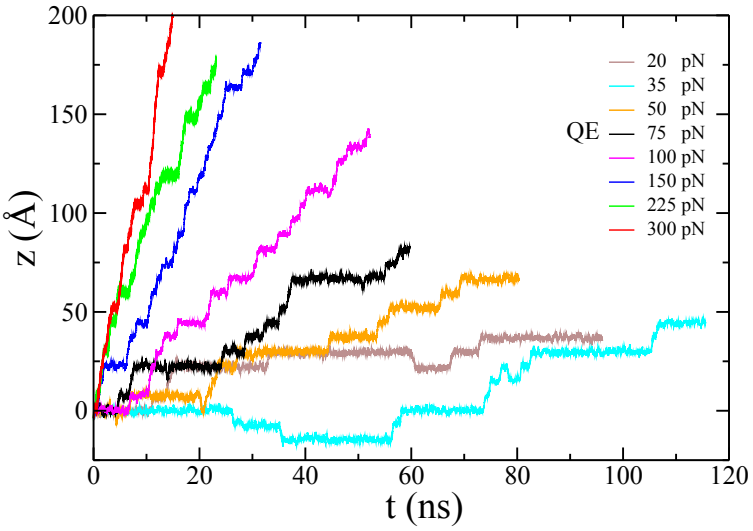

Figure S1: Electrically driven motion of ssDNA in a 1 M NaCl electrolyte in the DNA transistor. The biasing electric field varies from 6.25 to 93.75 mV/nm, corresponding to electric driving force QE changing from 20 to 300 pN.

Measurements of the friction coefficient of ssDNA in simulations

To obtain the friction coefficient of ssDNA in each electrolyte studied, we used the steered molecular dynamics (SMD) (1) method to pull ssDNA along the

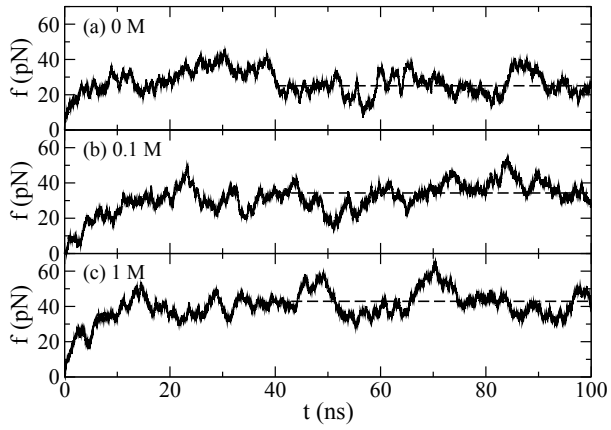

Figure S2: Driving forces on ssDNA when pulled by a harmonic spring (1 pN/Å). The pulling velocity is 1 nm/ns. The ion concentration is 0 M (a), 0.1 M (b) or 1 M (c). Each dashed line shows an average pulling force and the time span where the average was taken.

z-axis. As shown in Fig. 2b in the main paper, one end of a harmonic spring (k=1 pN/Å) is attached to the center of mass of all phosphorus atoms while the other end of spring is fixed on a stage that moves at a constant velocity v of 1 nm/ns. Figure S2 shows pulling forces f in the spring. ssDNA is at rest at the beginning of the simulation. As the pulling stage moves forward, the spring is stretched and the pulling force on ssDNA gradually increases. When ssDNA moves together with the pulling stage, a hydrodynamic force is exerted on the ssDNA molecule. When the mean velocity of ssDNA saturates at v, the pulling force $f = \xi v$, where ξ is the friction coefficient. From last 60-ns of the simulations for ssDNA in 0, 0.1 and 1 M NaCl electrolytes (see dashed lines in Fig. S2), the corresponding friction coefficients of ssDNA are 25.1 ± 5.9, 34.3 ± 7.0 and 42.9 ± 7.0 pN·ns/nm, respectively, where the error corresponds to the standard deviation around the mean spring force. Note that the standard deviation of the force can be computed as $\sqrt{kk_BT}$, resulting in 6.4 pN, which is consistent with errors in obtained friction coefficients.

Dependence of the ssDNA trapping force on ion concentrations

The mechanism of trapping (shown in Fig. 1 in the main paper) relies on the difference of one electron charge in two dielectric regions. Thus the trapping could be disrupted if one Na^+ binds a phosphate group inside electric trapping fields, reducing or even cancelling the trapping force. Thus, it is possible that the concentration of ions may affect the ssDNA trapping in the DNA transistor. However, when the ion concentration decreases to 0 M or increases to 1 M from 0.1 M (2), the force-position dependence changes little (see Fig. S3a and Fig. S3b).

In Fig. S3c, we show that the computed probability of a counterion residing on the DNA surface decays quickly with the residence time. Here, the residence time is defined as the time period when a counterion stays within 3 Å of any atom in ssDNA. The probability decreases quickly for short residence times (< 10 ps) that result from the frequent association and disassociation of a counterion with an ssDNA base. The mean residence time of a counterion on a phosphate group is longer and is typically about tens of picoseconds. The longest residence time of a counterion on a phosphate group found in our simulation is on the order of nanoseconds. The probability of a counterion residing in the vicinity of a phosphate group decreases with decreasing ion concentrations, in accord with a larger Debye length in the electrolyte with a lower ion concentration. As long as the residence time of a counterion on the DNA surface is much smaller than a typical time ($\sim d/v$, v the pulling velocity) for DNA to move out of the potential well, DNA is effectively trapped.

Note that the direction of each trapping field causes counterions to move away from the "dielectric-metal-dielectric" region (see Fig. 1b in paper), preventing counterions from residing on the ssDNA fragment inside the electric trapping fields.

Effective charge of ssDNA in a 1M NaCl electrolyte in the channel

We also measured the effective charge of ssDNA in a 1 M NaCl electrolyte confined in the same channel. Because of the reduced Debye length (3 Å), counterions closely surround the ssDNA molecule, yielding a stronger screening effect. Measured effective driving forces are smaller than those for ssDNA

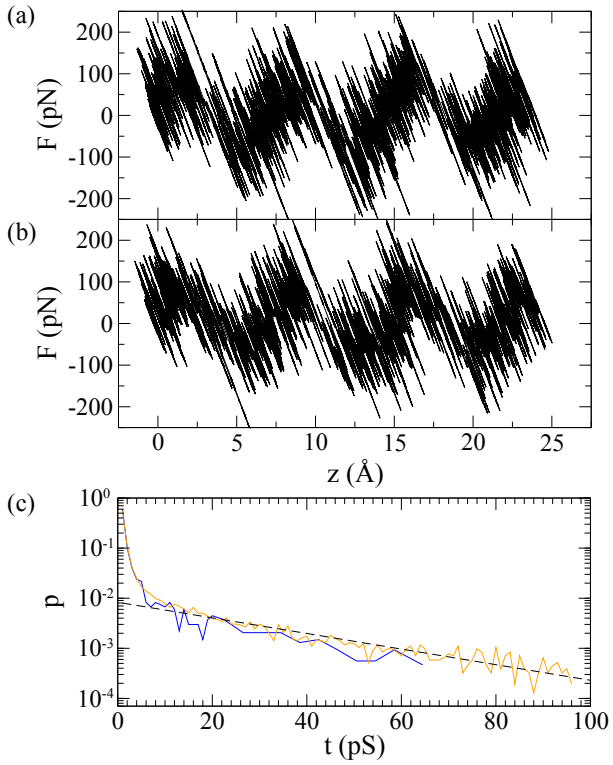

Figure S3: Effect of ion concentration on the ssDNA trapping force. (a,b) Force-position dependence of DNA pulled by a harmonic spring (100 pN/Å). The ion concentration is 0 M in (a) and 1.0 M in (b). (c) Probability of the residence time for a sodium ion residing on the ssDNA surface when ion concentrations are 0.1 M (blue) and 1.0 M (orange).

in a 0.1 M NaCl electrolyte (Fig. S4). Two spring constants were used to measure the effective electric driving forces. When k=1 pN/Å, the standard deviation of the spring forces resulting from thermal fluctuations is $(k_{\mathrm{B}}Tk)^{1/2}$, about 6.4 pN. Compared to the average force, force fluctuations are fairly big, which causes the measured spring forces in different electric fields to overlap significantly. Figure S4a shows results obtained using a weaker spring constant, 0.1 pN/Å. Because of smaller force fluctuations, measured forces in different electric fields are well separated. However, several hundreds of nano-seconds of simulation time are required. The mean spring force in each electric field is determined in the last 200 ns of simulation time.

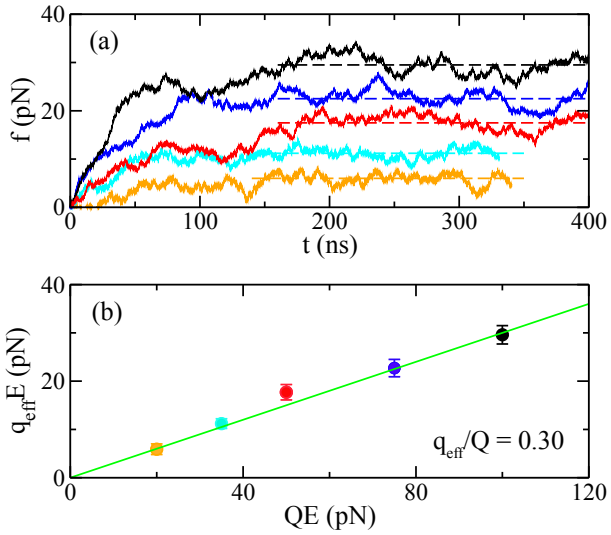

Figure S4: The effective charge q_{eff} of ssDNA in a 1 M NaCl electrolyte. (a) Time-dependent spring forces f that balance the effective electric driving forces $q_{\mathrm{eff}}E$ on ssDNA. The biasing electric field E= 6.25 (orange), 10.94 (cyan), 15.63 (red),23.44 (blue) and 28.13 (black) mV/nm. The spring constant is 0.1 pN/Å. Dashed lines show mean spring forces. (b) Effective electric driving force vs. electric driving force on bare ssDNA. The slope of the fitting line is the ratio between the effective charge q_{eff} of ssDNA and the charge Q of bare ssDNA. Error bars show standard deviations.

Thermostat in simulation

In our simulations, we applied a Langevin thermostat on atoms in the solid only, since the dissipative term ξv in the Langevin thermostate can apply a net frictional force on an atom or a molecule that moves at a none-zero mean veolcity $< v >$. To avoid this artifact, we choose to only thermostat constrained atoms in the solid. Physically, thermo-energy dissipates away through phonons (with a finite speed). Thus, it is physically more plausible to thermostat atoms near the boundary of a simulation system. Applying a thermostat to each atom would take out thermo-energy too fast.

References

1. Isralewitz, B., M. Gao, and K. Schulten, 2001. Steered molecular dynamics and mechanical functions of proteins. *Curr. Op. Struct. Biol.* 11:224–230.

2. Luan, B., H. Peng, S. Polonsky, S. Rossnagel, G. Stolovitzky, and G. Martyna, 2010. Base-by-base ratcheting of single stranded DNA through a solid-state nanopore. *Phys. Rev. Lett.* 104:238103.