

**Supplementary Information for**  
**Photothermally Assisted Thinning of Silicon Nitride Membranes for Ultrathin Asymmetric**  
**Nanopores**

By

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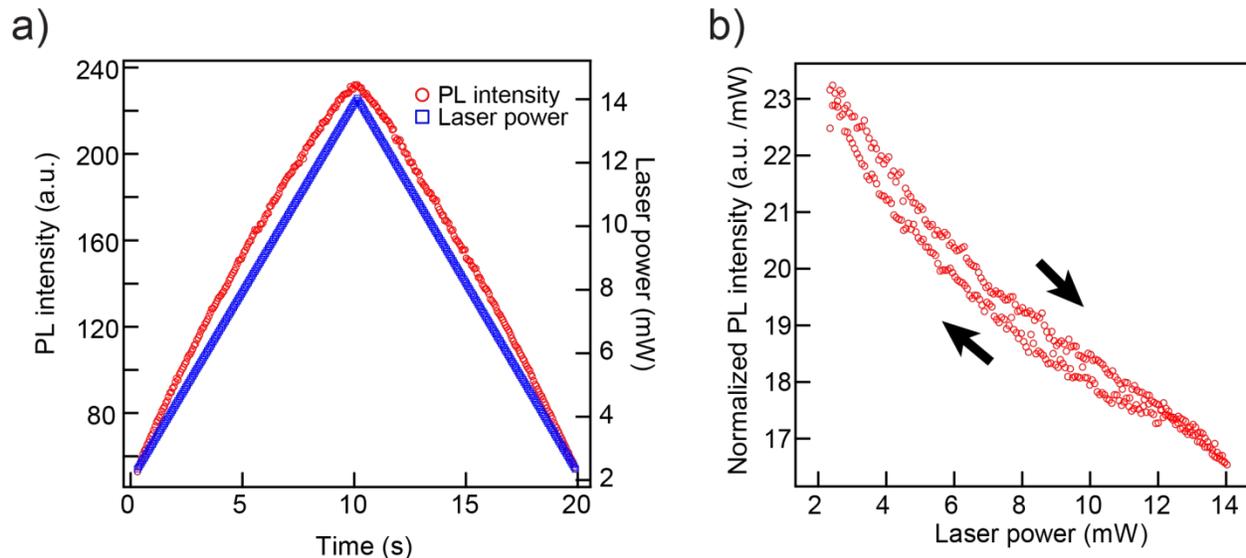
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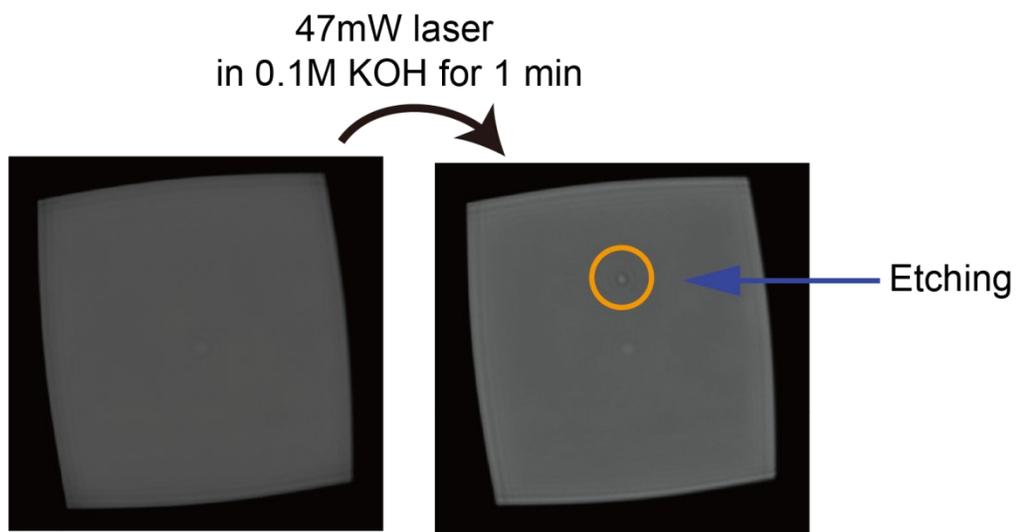
Phone: +1-617-373-7412

## 1. PL kinetics of SiN<sub>x</sub> membrane excited by 532 nm laser



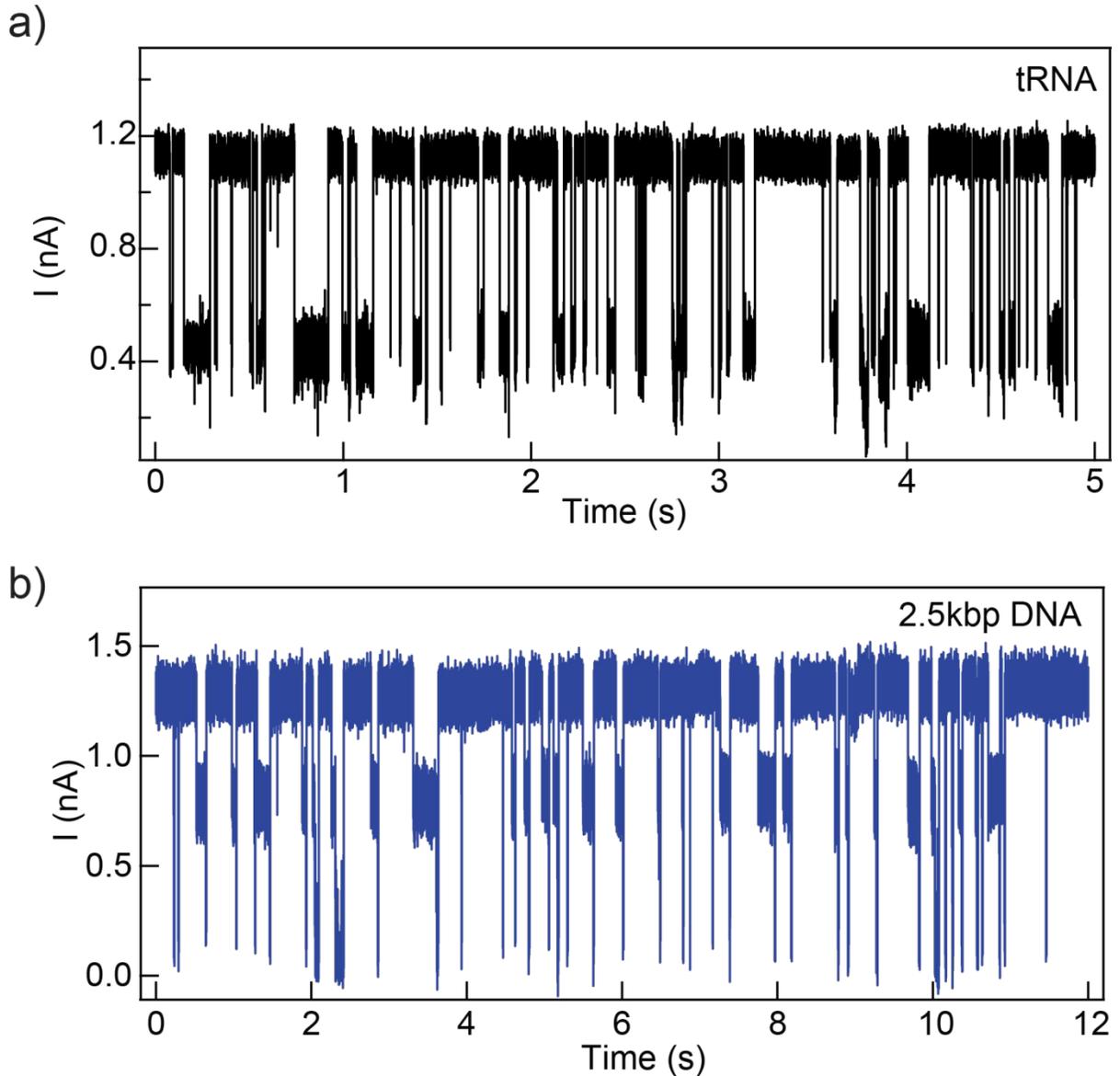
**Figure S1.** (a) Continuous PL trace (red) and Laser power profile (blue) during sweeping laser power. Measurement was demonstrated by focusing 532 nm laser on 75 nm SiN<sub>x</sub> membrane and ramping-up/-down the laser power, ranging from 2.4 mW to 14 mW, in 10 sec. PL passed through two long pass filter ( $> 532$  nm) was detected using a monochrome CMOS camera (ASI290MM, ZWO) at a framerate of 16 Hz. PL intensity values were obtained by averaging the intensity over the centroid spot in a diameter of 15 pixels, which corresponds to 750 nm. (b) Normalized PL intensity vs. Laser power. Normalized PL intensity was calculated by dividing the PL intensity by the laser power, based on the data shown in panel a. Arrows represent time series of PL intensity at each laser power. Normalized PL intensity decays with laser power, representing temperature dependence on SiN<sub>x</sub> PL efficiency (25% decrease in PL efficiency over a  $\Delta T \sim 64$  K, based on COMSOL). The minor hysteresis in **Figure S1b** represents minor SiN<sub>x</sub> etching during laser power sweep, yet the trend is reproducible.

2. SiN<sub>x</sub> etching in 0.1 M KOH induced by photothermal heating



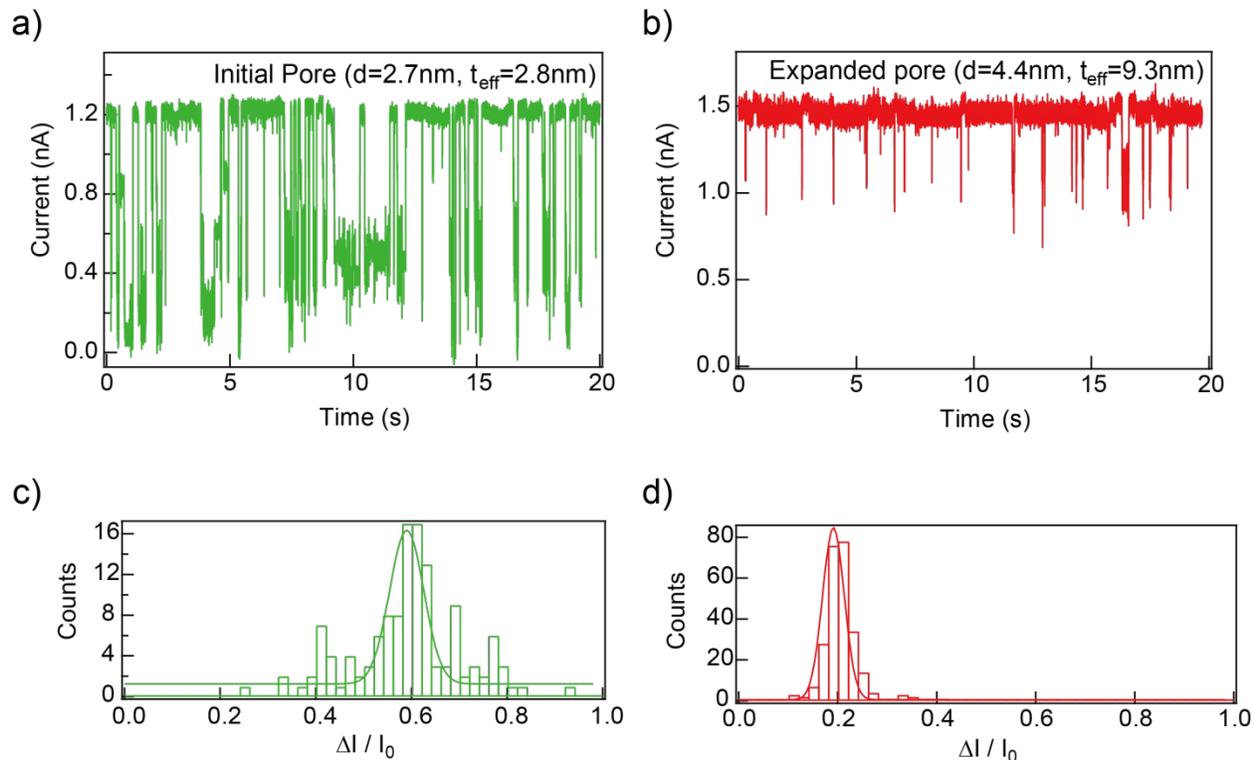
**Figure S2.** Optical image before and after 47mW laser in 0.1 M KOH for 1 min. Orange circular on right highlights the etched location after laser illumination.

### 3. Continuous current traces of DNA and tRNA translocation



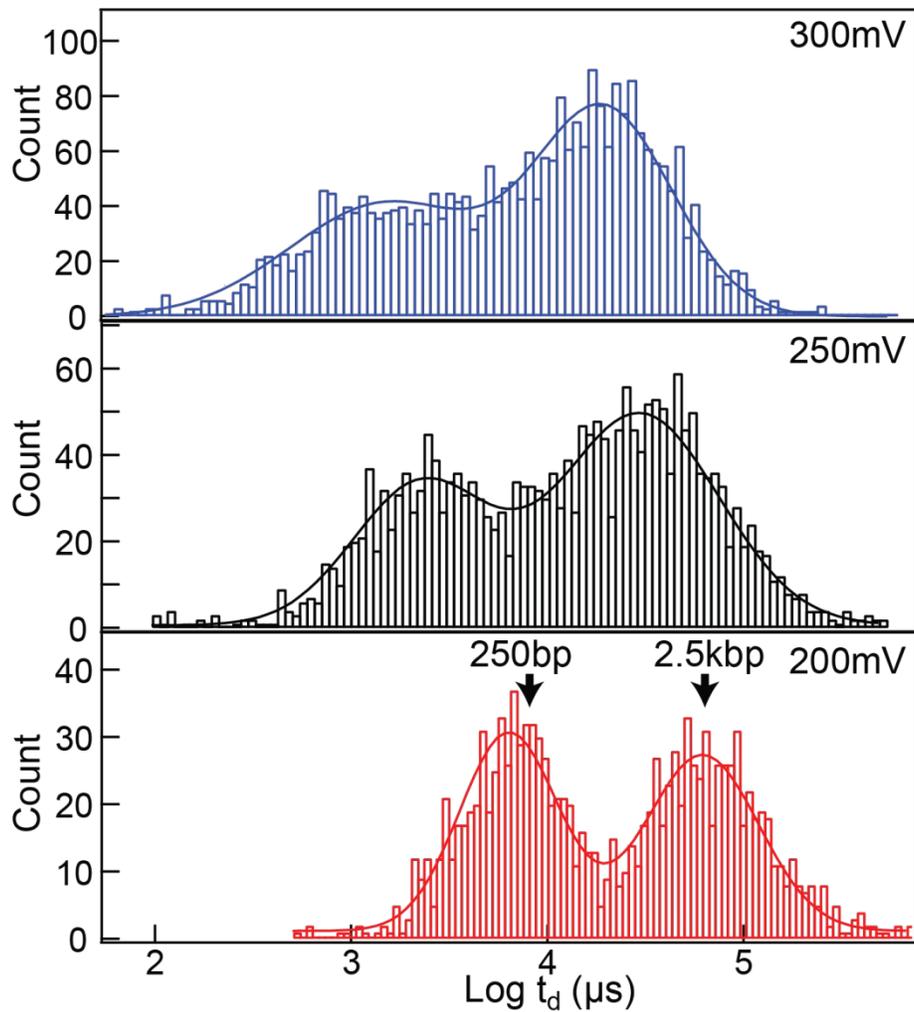
**Figure S3.** Current vs. time traces of (a) tRNA arginine translocation through 2.5 nm diameter pore with 5.6 nm thickness at 300 mV. (b) 2.5 kbp DNA translocation through 2.4 nm diameter pore with 3 nm thickness at 300 mV. All traces were obtained using 0.4 M KCl. Note that although current spikes appear to be below 0 nA, these are due to capacitive noise, and all mean blocked levels were  $>0$  nA despite appearance, due to noise in the blocked level.

#### 4. Relation between pore size and thickness



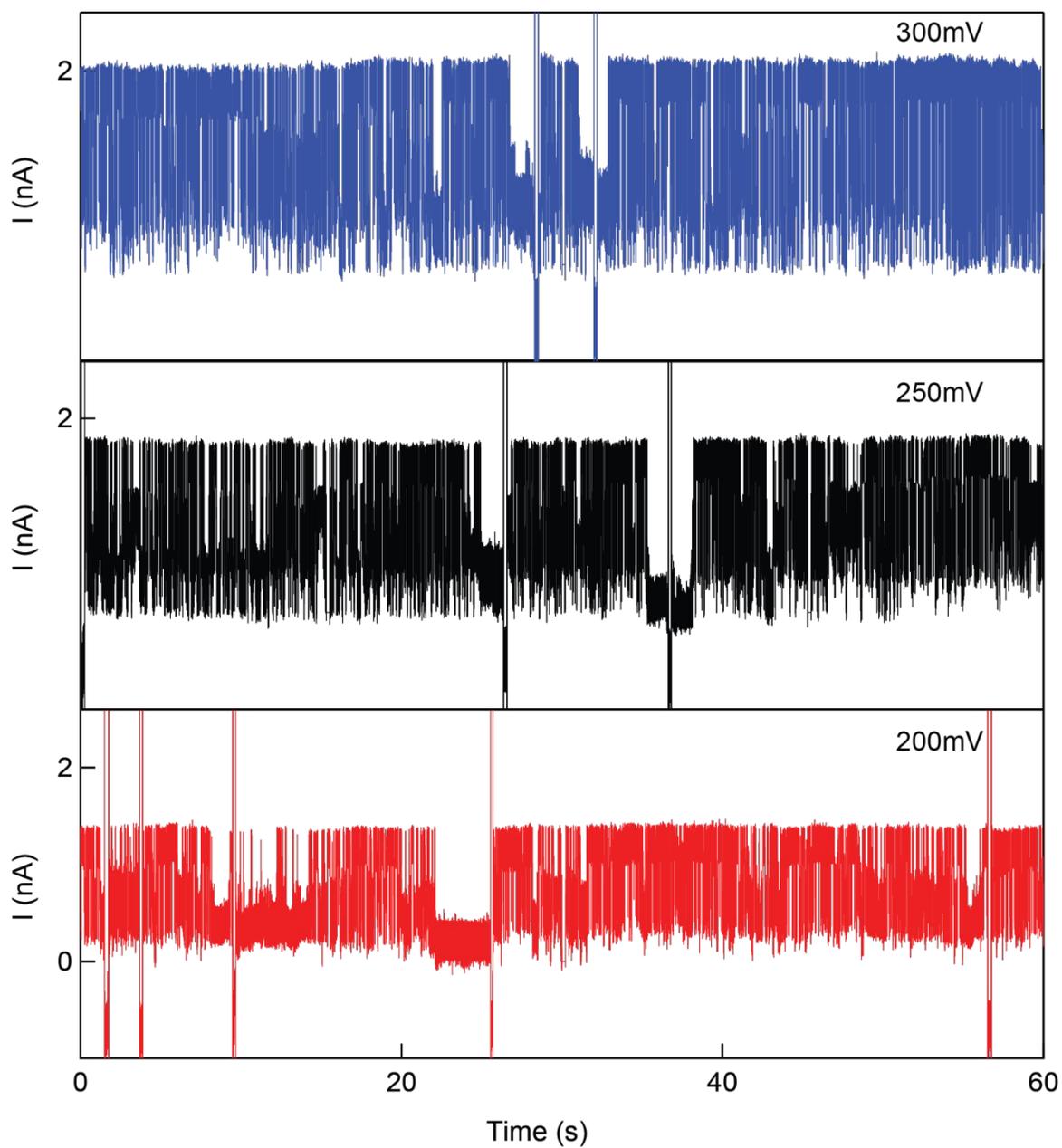
**Figure S4.** Relation between pore size and effective thickness for a gradually expanding pore. (a) and (b) 20-sec current trace of 2.5 kbp DNA at 300mV before and after pore expansion. (c) and (d) histogram of  $\Delta I / I_0$  before and after pore expansion. Mean  $\Delta I / I_0$  before and after expansion are 0.59 and 0.19, respectively.

5. Histograms of  $\text{Log } t_d$  for mixture of 250 bp and 2.5 kbp



**Figure S5.** Histograms of  $\text{log } t_d$  at 200, 250 and 300 mV. Double Gaussian fits were applied to obtain mean values of both peaks. Mean  $\text{log } t_d$  values for 250 bp were  $3.80 \pm 0.01$ ,  $3.36 \pm 0.03$  and  $3.19 \pm 0.05 \mu\text{s}$ . Mean  $\text{log } t_d$  values for 2.5 kbp were  $4.79 \pm 0.01$ ,  $4.47 \pm 0.02$  and  $4.29 \pm 0.02 \mu\text{s}$ .

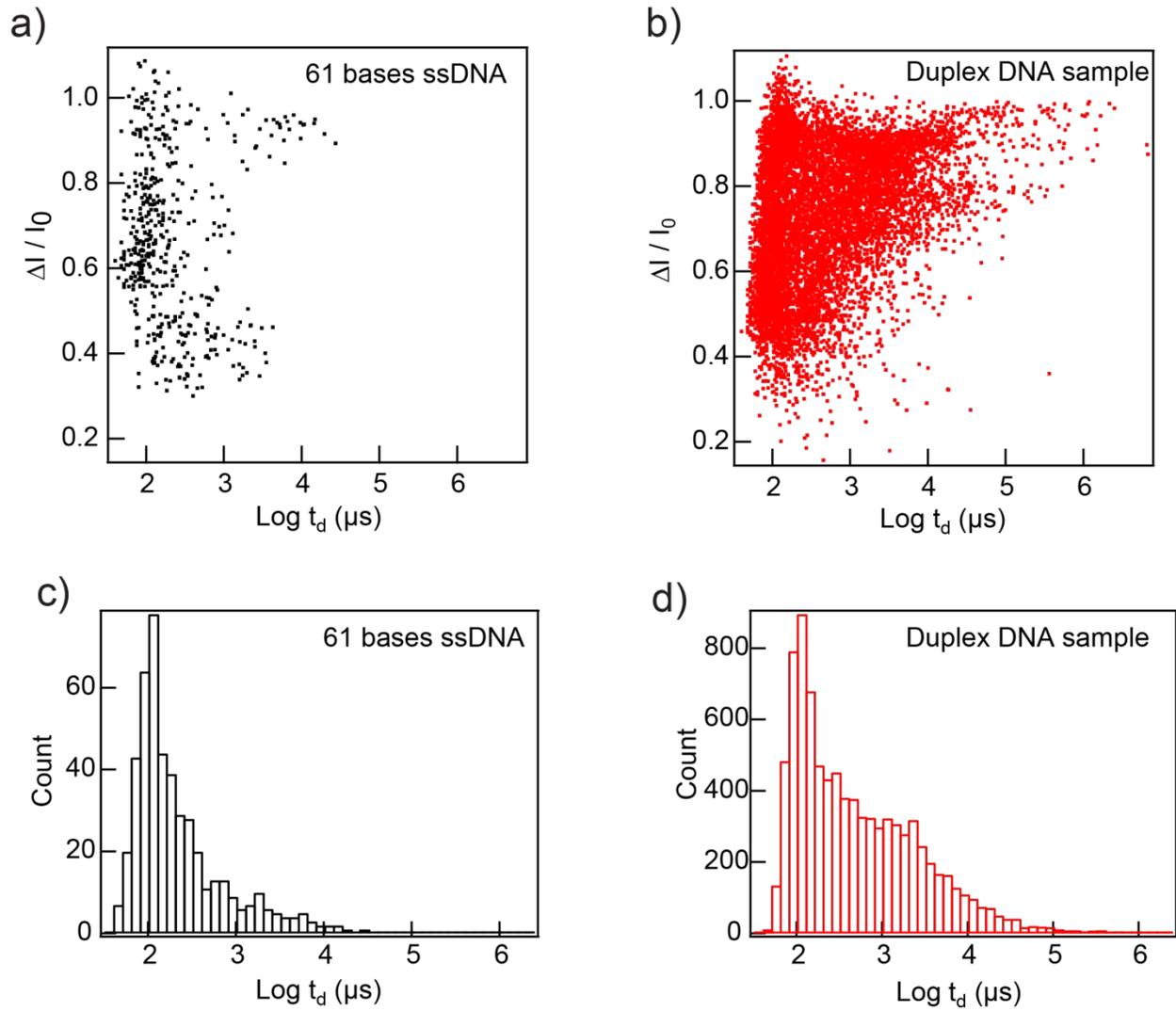
6. Continuous current traces for a mixture of 250 bp and 2.5 kbp dsDNA.



**Figure S6.** 60 s continuous current traces at 200, 250 and 300 mV for 30 nM 250 bp and 3 nM 2.5 kbp mixture.

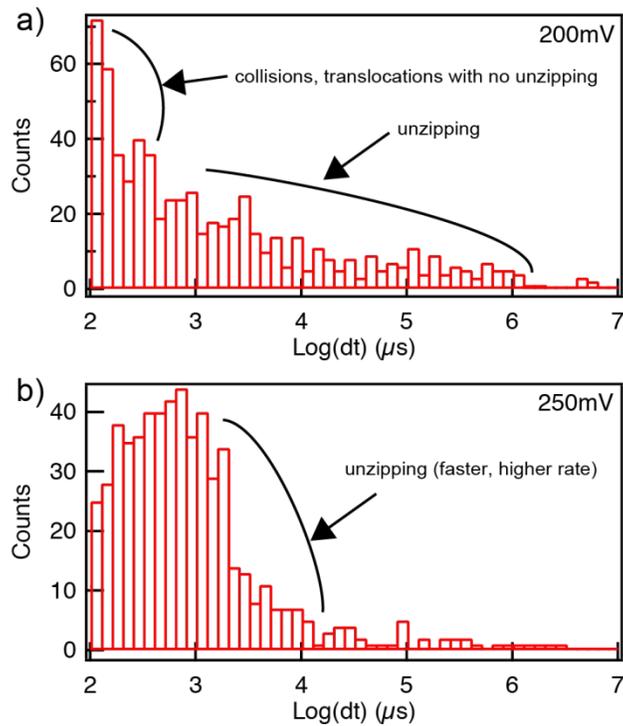


8. Scatter plots and log  $t_d$  histogram of 61-nucleotide ssDNA and duplex DNA samples



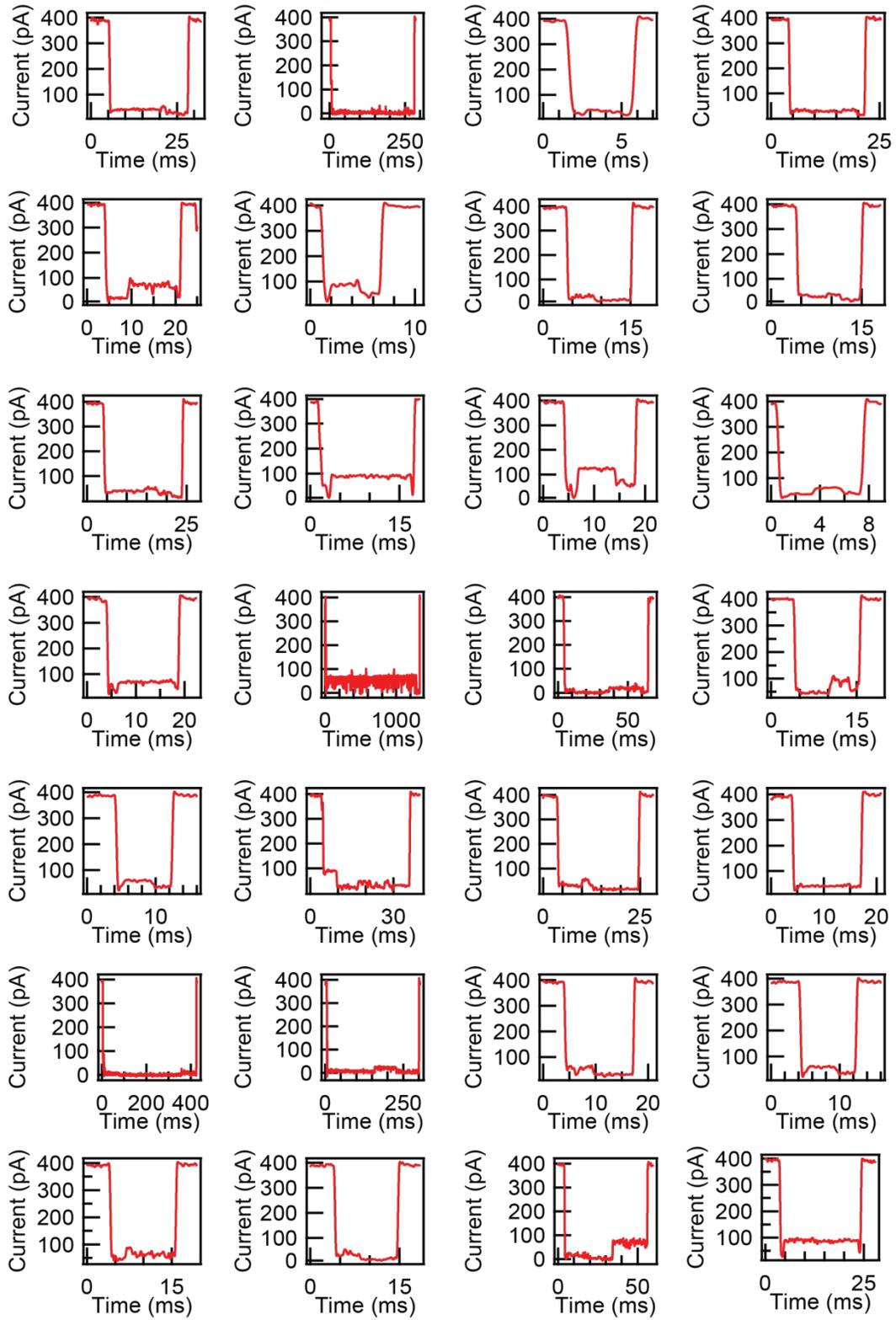
**Figure S8.** (a) Scatter plots of 61-nucleotide ssDNA translocation through 1.4 nm pore with 1.8 nm thickness at 150 mV ( $n = 476$ ). (b) Scatter plots of duplex DNA sample through the same pore at 200 mV ( $n = 2,967$ ). (c) Histogram of 61-nucleotide ssDNA log  $t_d$  (d) Histogram of duplex DNA sample log  $t_d$ .

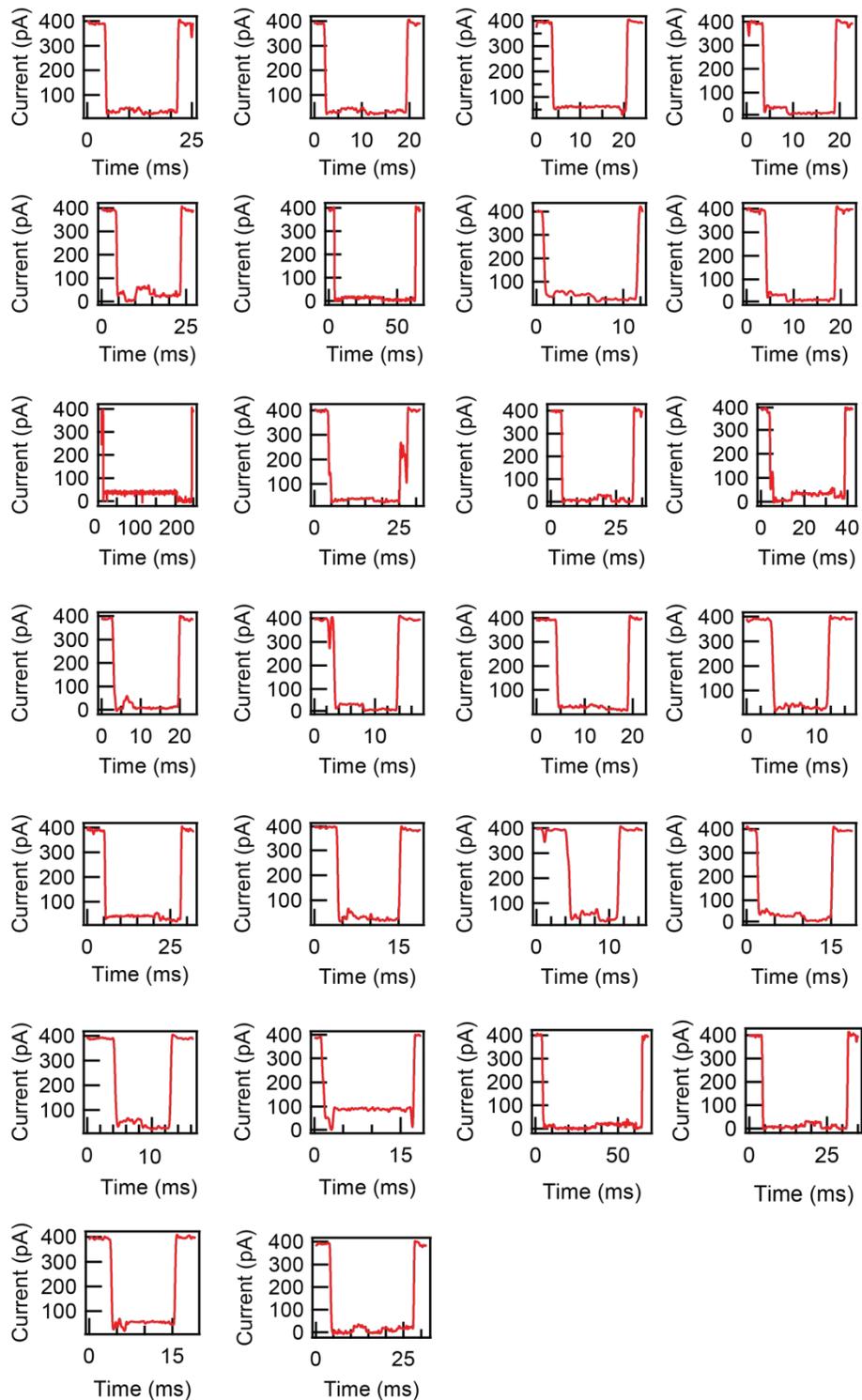
## 9. Effect of voltage on dsDNA unzipping



**Figure S9.** Dwell-time histograms of dsDNA unzipping using 1.3 nm pore with 2 nm effective thickness at 200 and 250 mV. When voltage increases, characteristic dwell-time that corresponds to dsDNA unzipping is reduced by an order of magnitude.

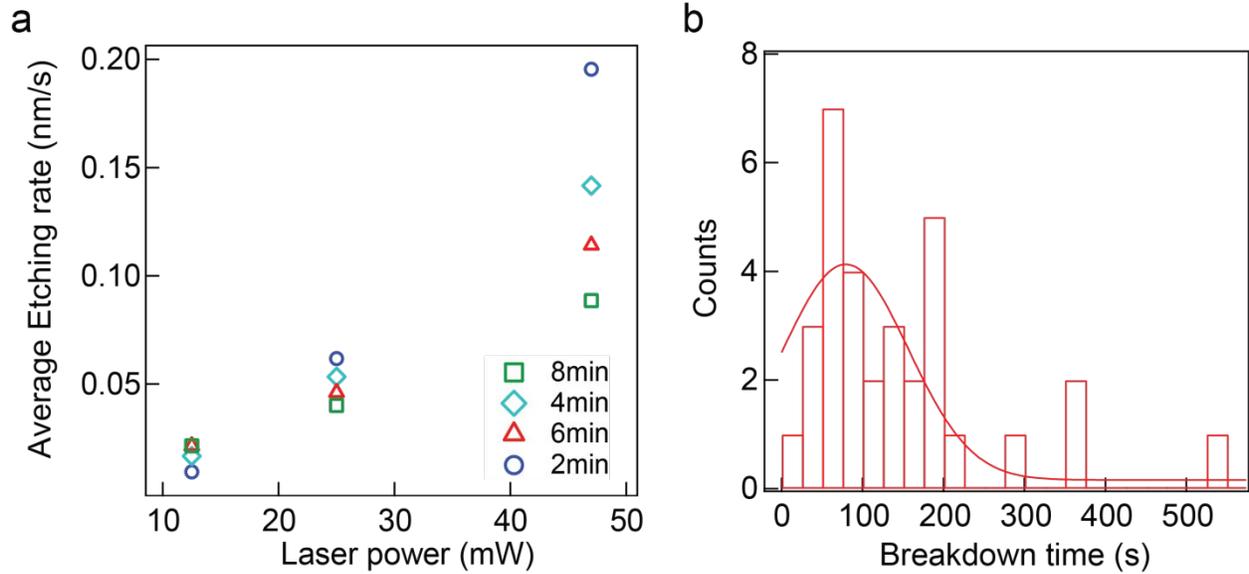
10. Random poly (dC) and poly(dA) discrimination current traces, in which many of the traces show 1-2-1 two-level signals.





**Figure S10.** Random current traces ( $n = 54$ ) which show discrimination between levels of poly(dC) and poly(dA) from the experiment of Figure 6. Deep→shallow→deep events are not always seen (only in 18.6% of the long dwell data), presumably due to some molecules not having all three oligos hybridized to the template DNA, or some of the molecules not entering the pore in an unzippable conformation (data low-pass filtered to 1kHz).

## 11. Average SiN<sub>x</sub> etching rate and breakdown time



**Figure S11.** (a) Average etch rate vs. laser power. Average etch rates were calculated using etch depths from measurements shown in Figure 3b. Etch rate depends on laser power and etching time, representing etching rate decreases by time due to less heating of thin membrane. The average etching rate of four etching times at  $P = 12.5, 25$  and  $45$  mW are  $0.017, 0.050$  and  $0.135$  nm/s, respectively. (b) Histogram of all breakdown times using  $75$  nm thick SiN<sub>x</sub> membrane under  $P = 47$  mW and  $V = 1$  V ( $N=39$ ). Average breakdown and standard deviation are  $79.268 \pm 16.1$  s and  $109.42 \pm 27$  s, representing etch rates variation of up to a factor of 2.

## 12. Summary of tested chips

Table S1 Chip information 1

	Total tried chips (105)	Chips for Pore fabrication Chips (49)	Usable pore Chips (35)
Wafer A	62	22	15
Wafer B	43	27	20

Table S2 Chip information 2

	Overshot chips	Unusable pore (over-etched pore)	Desired pore (< 4 nm)
Wafer A	5 of 62	7 of 22 (4 of 22)	11 of 15
Wafer B	5 of 43	7 of 27 (0 of 27)	18 of 20

Table S1 and S2 summarize chip information for etching and pore fabrication. When fabricating a pore using our method, pores that were unclean or over-etched are referred to as unusable pores. The yield of the usable pores in our method was 71.4 % (35/49 chips). Among clean chips that have stable baseline current signals, the yield of desired pores for high-sensitivity DNA detection (< 4 nm diameter) was 82.9 % (29/35 chips). The 35 chip count includes 29 chips whose geometric parameters are shown in Figure 4b, while 3 chips were fabricated using a blue laser, and another 3 chips were used for pore formation in 1 M KCl. Less than 10 % of the total tested chips were overshoot in size or too overetched such that the membrane did not remain, respectively (10/105 and 4/49 chips).