Supporting Information for:

Experimental Study on Single Biomolecules Sensing using MoS_2-Graphene Heterostructure Nanopores

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Protein excluded volume model: Since the thickness of the heterostructure is very thin, it is more accurate to take the access regions on both side of the nanopore into account. Therefore, we use protein excluded volume model[1] to estimate the range of blockage current of BSA:

$$\Delta I = I_0 \frac{\Lambda(t)}{H_{eff} A_p}$$

Here ΔI is blockage current, I_0 is open pore current, $\Lambda(t)$ is the instantaneous volume of BSA molecules, H_{eff} is the effective length of nanopore which include the access regions as $H_{eff} = L + \frac{d\pi}{4}$, A_p is the area of nanopore. To make sure BSA molecules are totally translocated through the nanopore, we take $\Lambda(t)$ as the widest part of BSA along the long axis, which is 4 nm. The calculation results show that only the signals in small pores with $\Delta I \geq$~2.3 nA can be counted as effective.

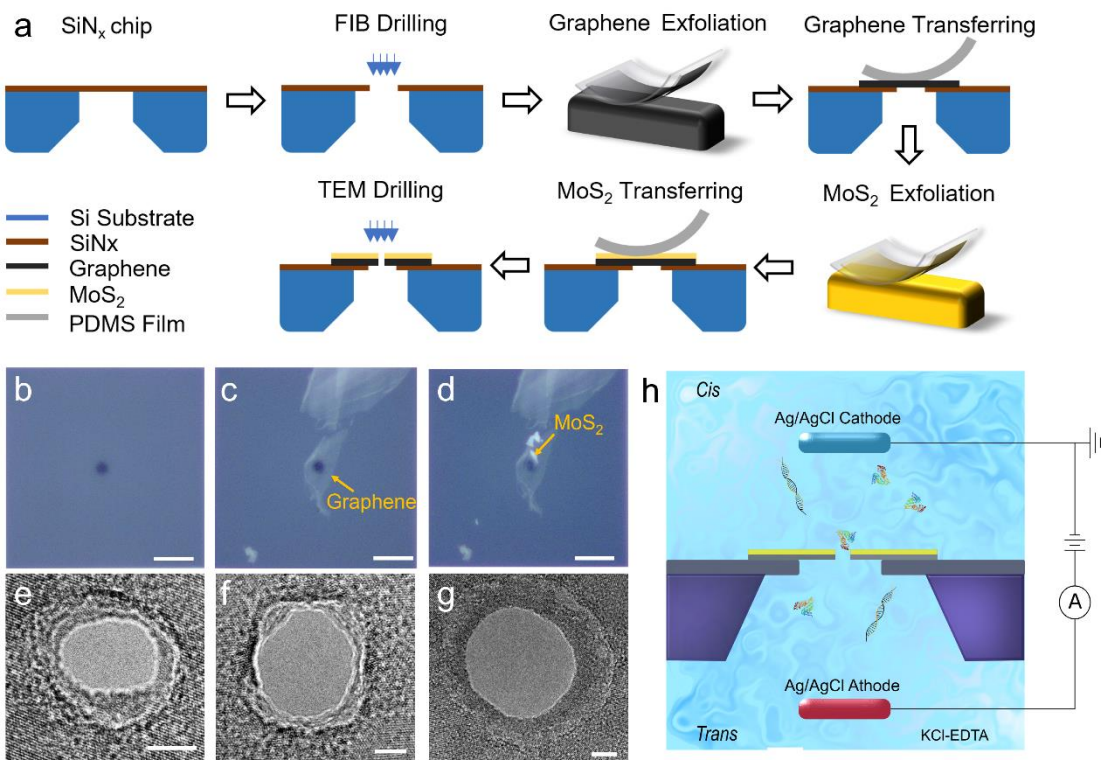

Fig. S1 (a) Schematic diagram of fabrication of the heterostructure nanopore. (b) An FIB pore in diameter of 1 μm on the SiNx membrane, (c) an exfoliated graphene membrane transferred onto the FIB pore and (d) an exfoliated MoS2 membrane transferred onto the graphene membrane and covering the FIB pore under the optical microscope (scale bar: 10 μm). (e)-(g) TEM pictures of heterostructure nanopores with diameter of ~5 nm, ~15 nm and ~20 nm (scale bar: 5 nm). (h) A schematic diagram of the heterostructure nanopore device and its test system.

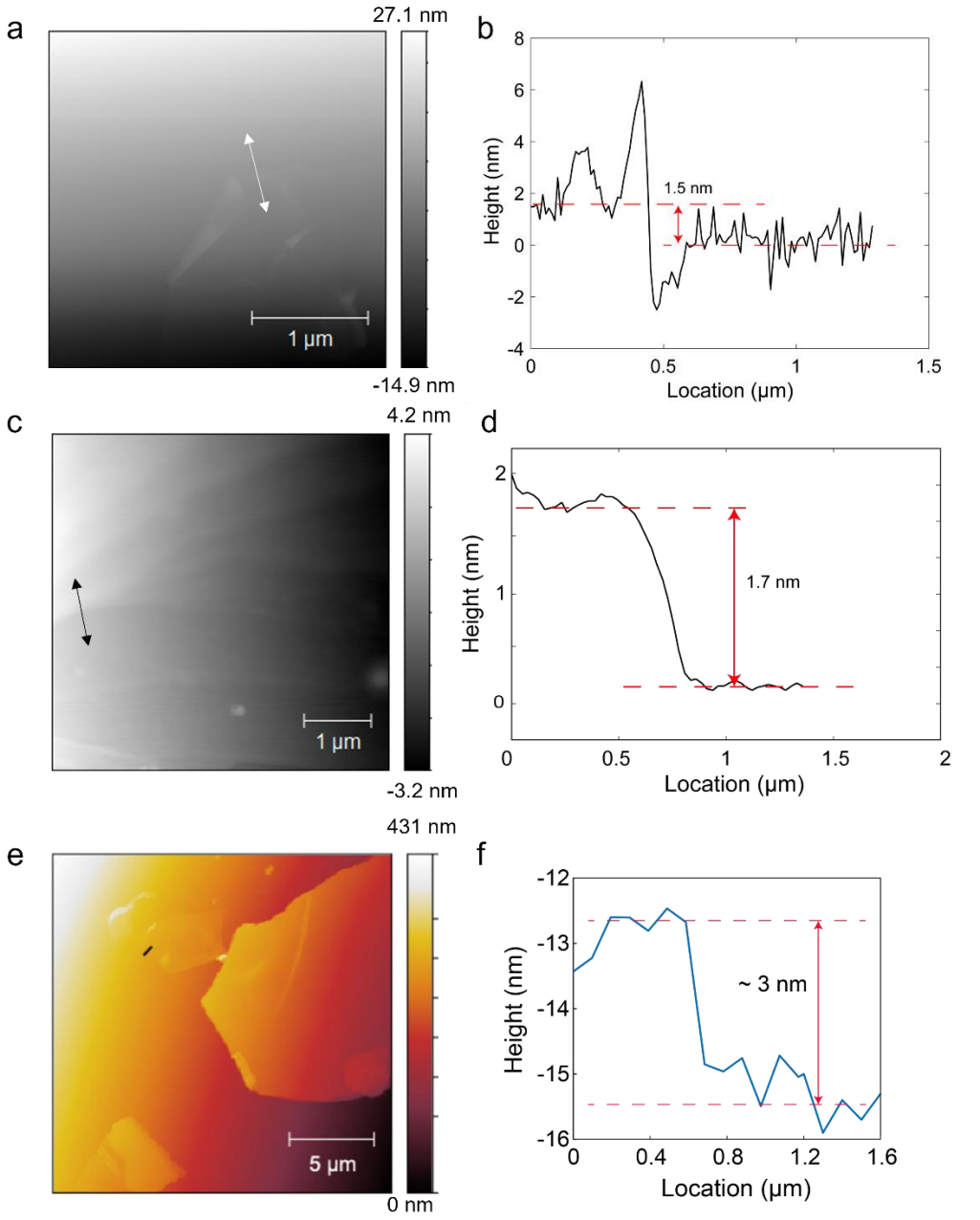

Fig. S2 Atomic Force Microscope scan for the thickness of (a) a graphene sample, (c) a MoS_2 sample and (e) a heterostructure sample. (b), (d), (f) are height variation across the specific locations (marked with arrow and short strip).

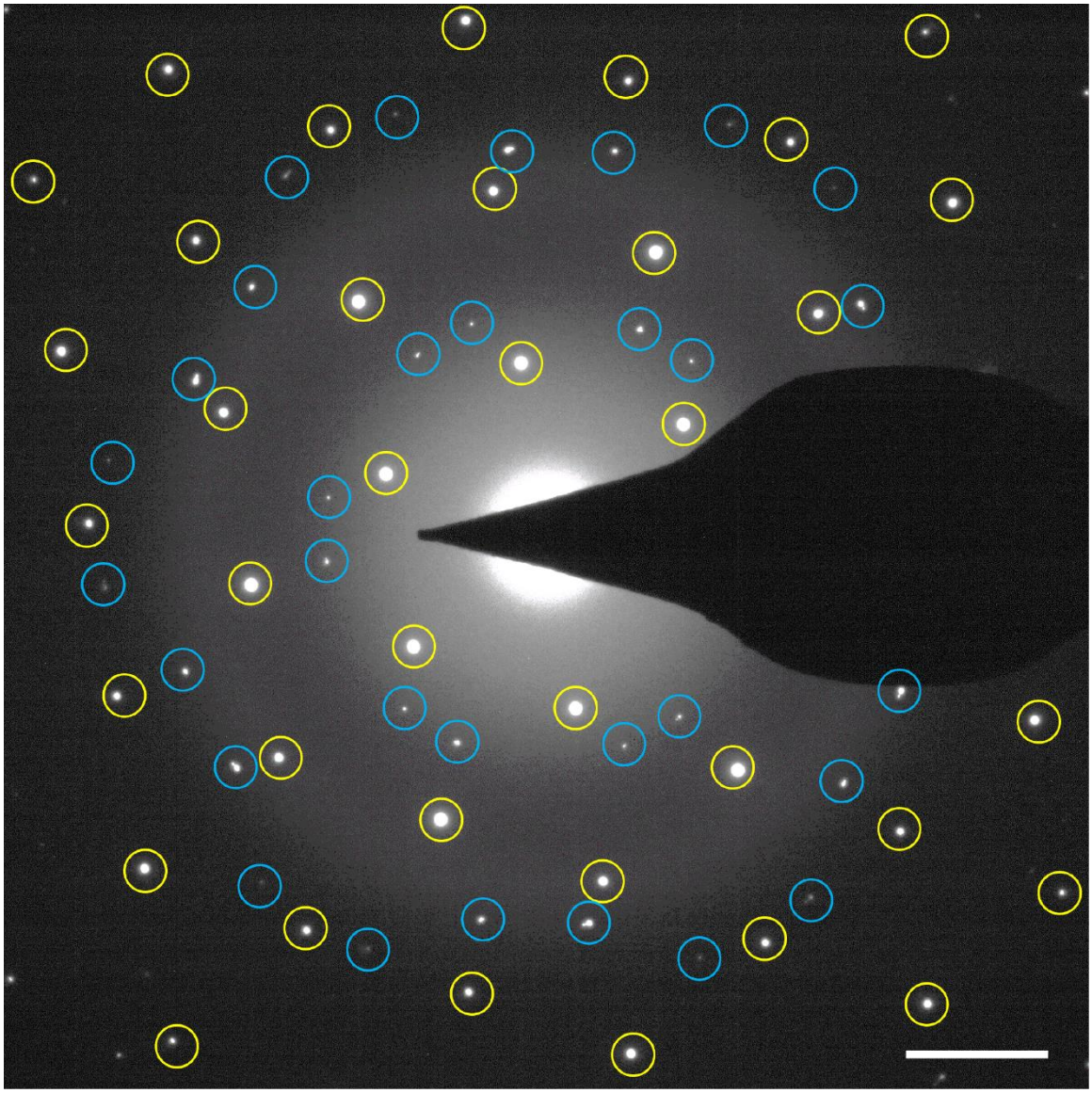

Fig. S3 Electronic diffraction of MoS_2-Graphene heterostructure. There are two sets of diffraction spots of MoS_2 (yellow circle) and Graphene (blue circle). The ratio of the two lattice constants (a(Gr)/a(MoS2)) obtained through experiments is about 0.78, while the lattice parameters of Graphene and MoS2 previously reported in the literature are 2.46 Å[1] and 3.19 Å,[2] and the ratio of the two is 0.778, which is consistent with our experimental data. The scale bar in the picture is 3 nm.

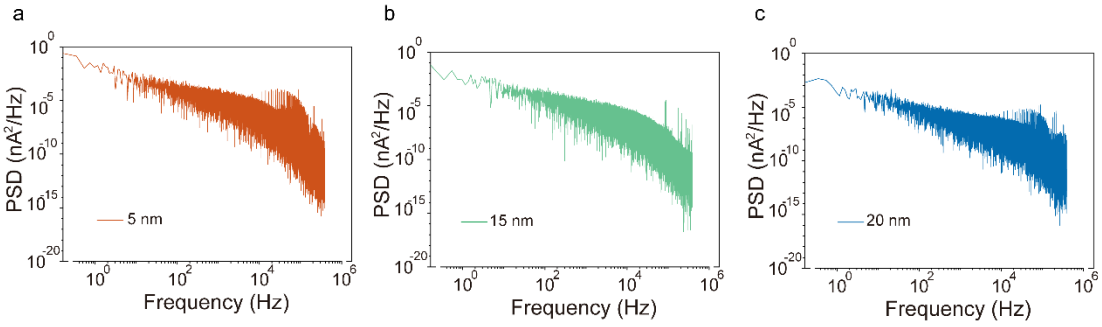

Fig. S4 PSD analysis of three nanopores with different pore sizes at 400 mV.

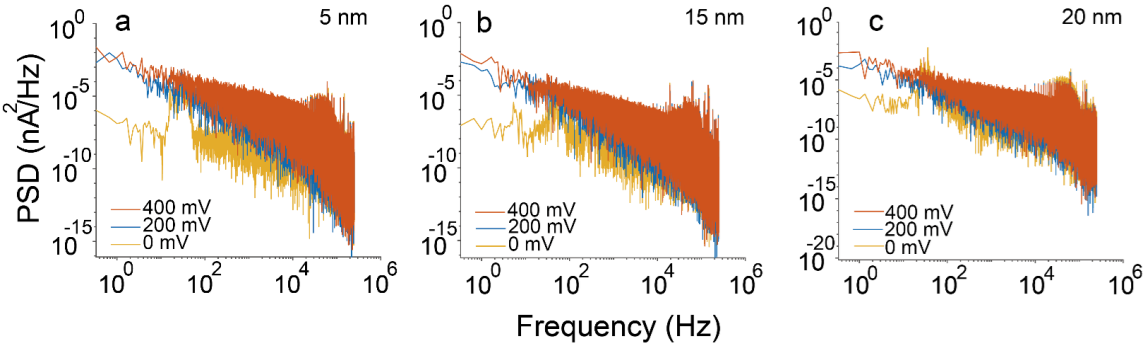

Fig. S5 PSD at different bias. Typical power spectrum density analysis of three nanopore devices with different pore size. At 0 mV, 200 mV and 400 mV bias, regardless of the pore size, as the bias increases, the 1/*f* noise increases too.

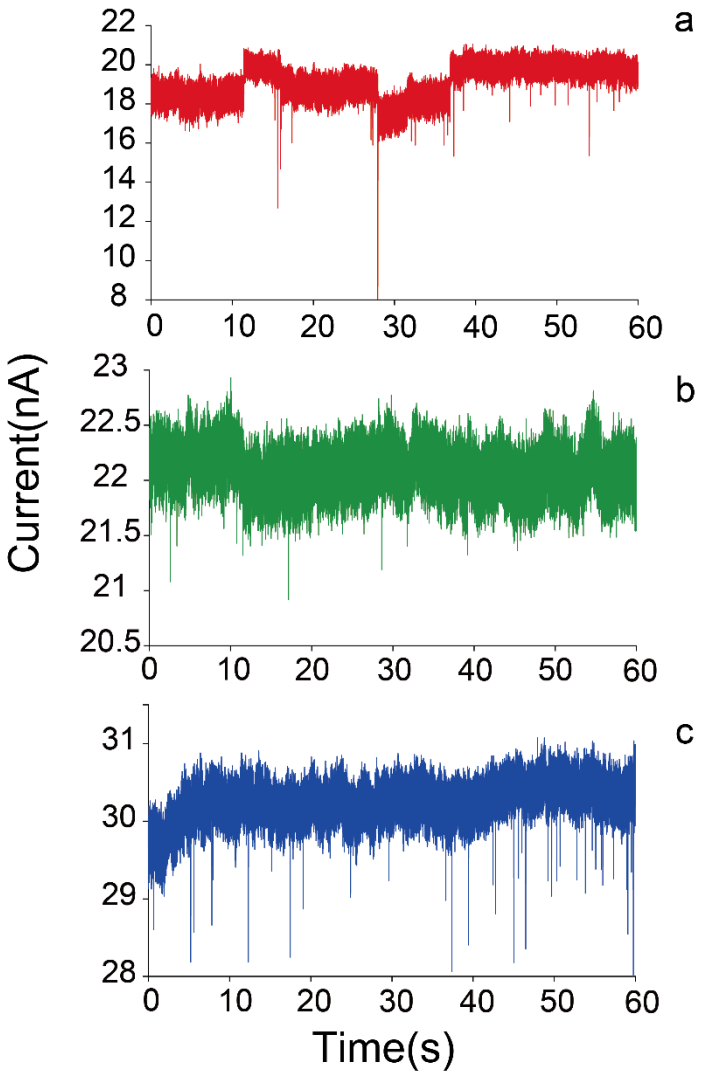

Fig. S6 DNA traces of three nanopores with diameter of 7 nm, 10 nm and 14 nm from (a) to (c) at 400 mV.

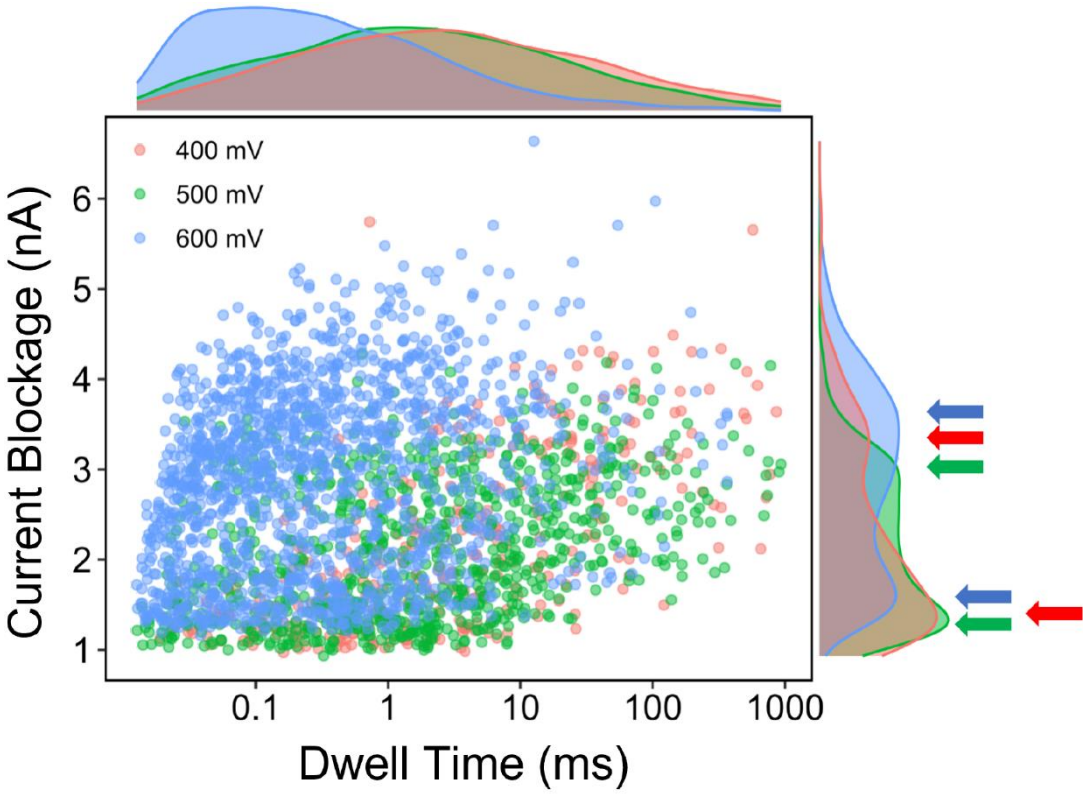

Fig. S7 Scatter plot of device 1. Device 1 has a diameter of 6.75 nm. At every bias, the BSA translocation signals have two clusters, reflected by two peaks in current blockage density distribution. The first cluster is possibly the group of signals that BSA molecules are not fully translocated.[3] Therefore, these signals should be dismissed during the analysis.

Table. S1 The BSA translocation details of 6 heterostructure nanopores with various geometric parameters and bias voltage ranges.

Device ID	Nanopore Diameter	Signals Number	Signals Number/Ratio (DTa) > 10 ms)	Signals Number/Ratio (DTa) > 100 ms)	Bias Range (mV)
1	6.75 nm	296	81/27.4%	24/8.1%	400-600
2	5.09 nm	215	21/9.8%	3/1.4%	400-600
3	5.74 nm	180	24/13.3%	1/0.6%	400-600
4	15.62 nm	3399	93/2.7%	0/0%	200-400
5	15.72 nm	27301	52/0.2%	0/0%	100-400
6	19.96 nm	11003	68/0.6%	4/0.04%	100-300

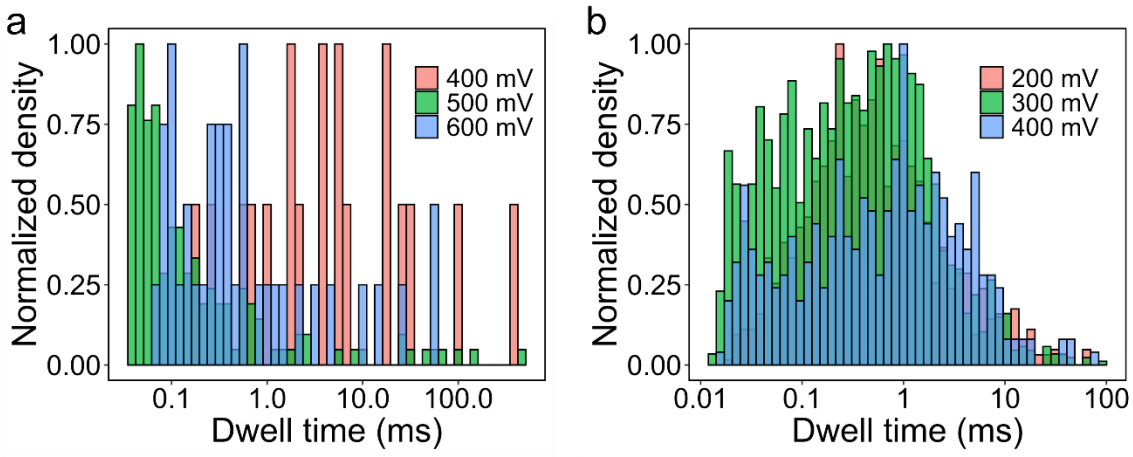

Fig. S8 The dwell time distribution of device 2 and device 4.

Table. S2 The detail information of BSA translocation through SiNx nanopores

ID	Diameter	Signals Number	Signals Number/Ratio (DT>10 ms)	Signals Number/Ratio (DT>100 ms)	Bias Voltage Range(mV)
1	9 nm	6291	16/0.25%	0/0	500-600
2	25 nm	6611	36/0.54%	0/0	100-400
3	12 nm	23778	56/0.34%	0/0	300-500
4	14 nm	15740	2/0.01%	0/0	100-400

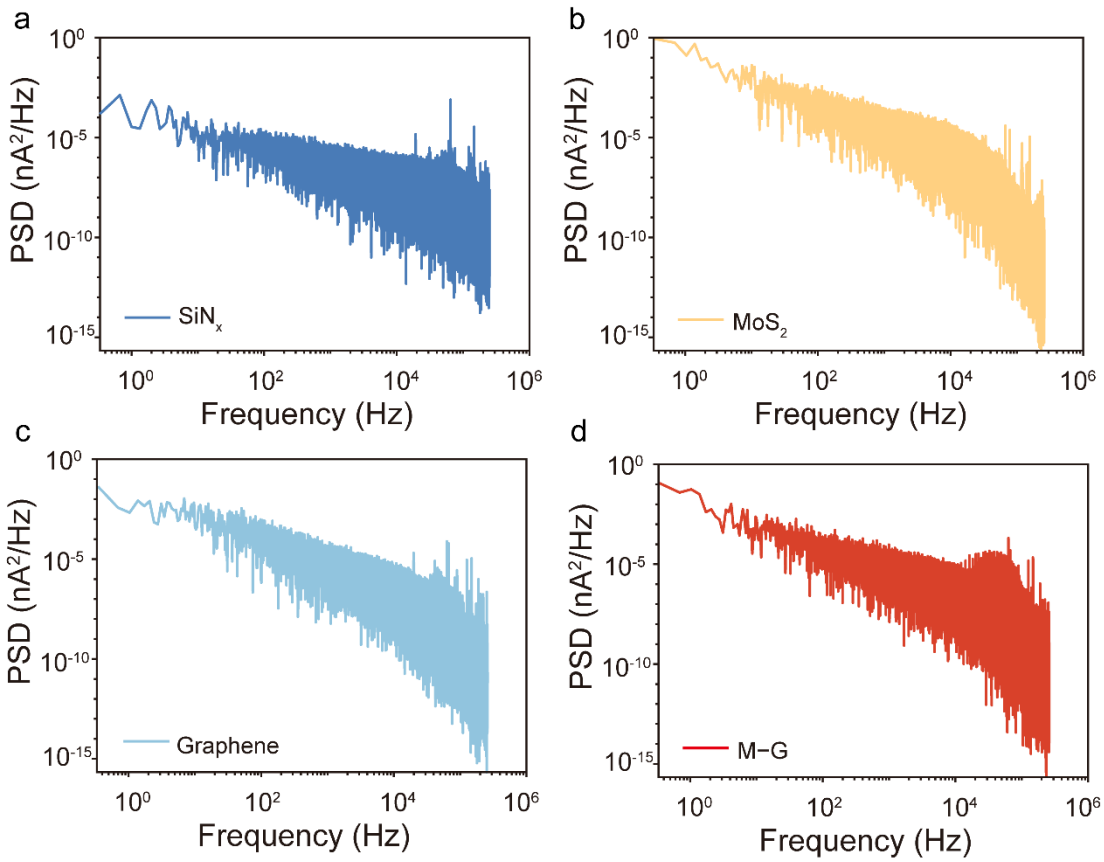

Fig. S9 PSD of four types of nanopores with diameter of ~7 nm at 400 mV.

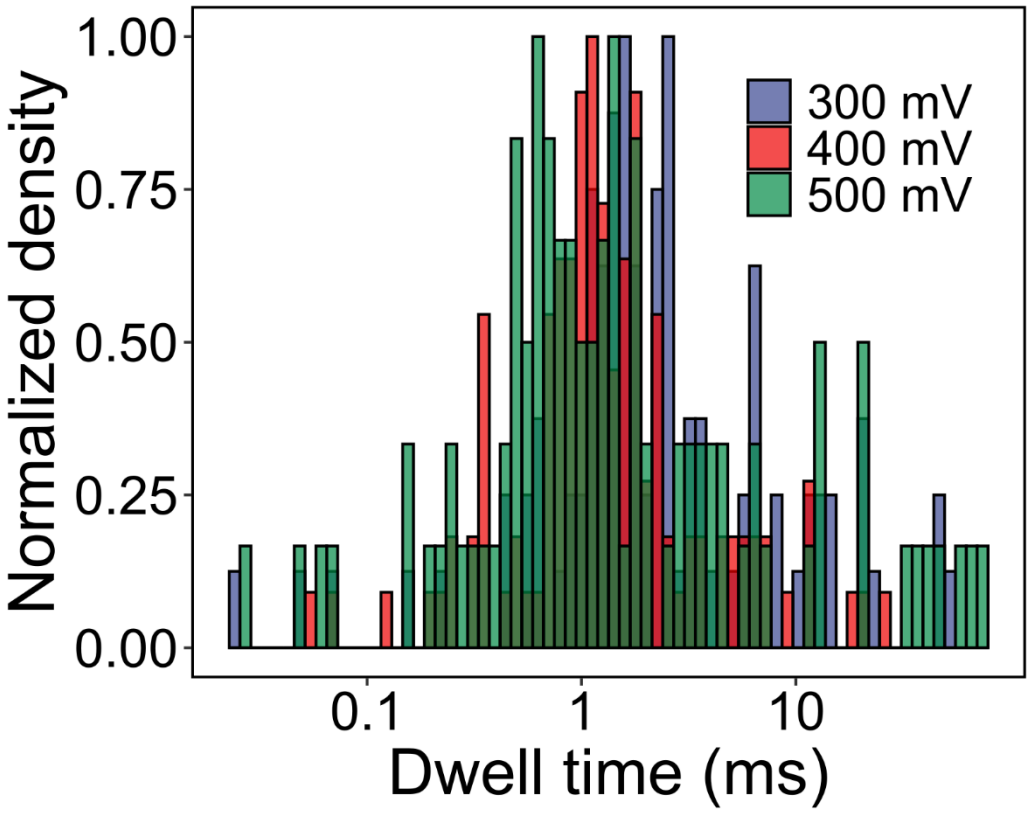

Fig. S10 DNA translocation dwell time histogram of SiN_x nanopore with diameter of ~15 nm

References

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2 Y. Baskin and L. Meyer, *Phys. Rev.*, 1955, 100, 544–544.

3 Z. Y. Zhu, Y. C. Cheng and U. Schwingenschlögl, *Phys. Rev. B*, 2011, 84, 153402.

4 D. Fologea, B. Ledden, D. S. McNabb and J. Li, *Appl. Phys. Lett.*, 2007, 91, 053901.