Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Electric Field Interference and Bimodal Particle Translocation in Nano-Integrated Multipores

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Supplementary Information includes:

1. Supplementary Figures (Figs. S1-S12)

2. Supplementary references

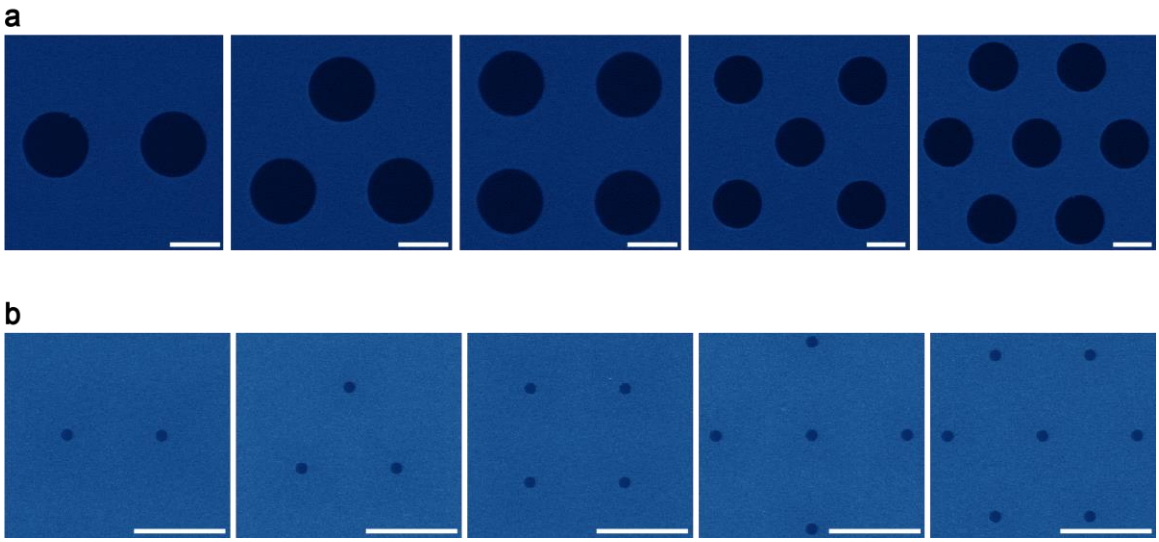

Figure S1. **a-b,** False-colored scanning electron microscopy images of multipores with inter-pore distance D_{ch} of 1 μm (a) and 10 μm (b). Bars denote 1 μm and 10 μm, respectively, in (a) and (b).

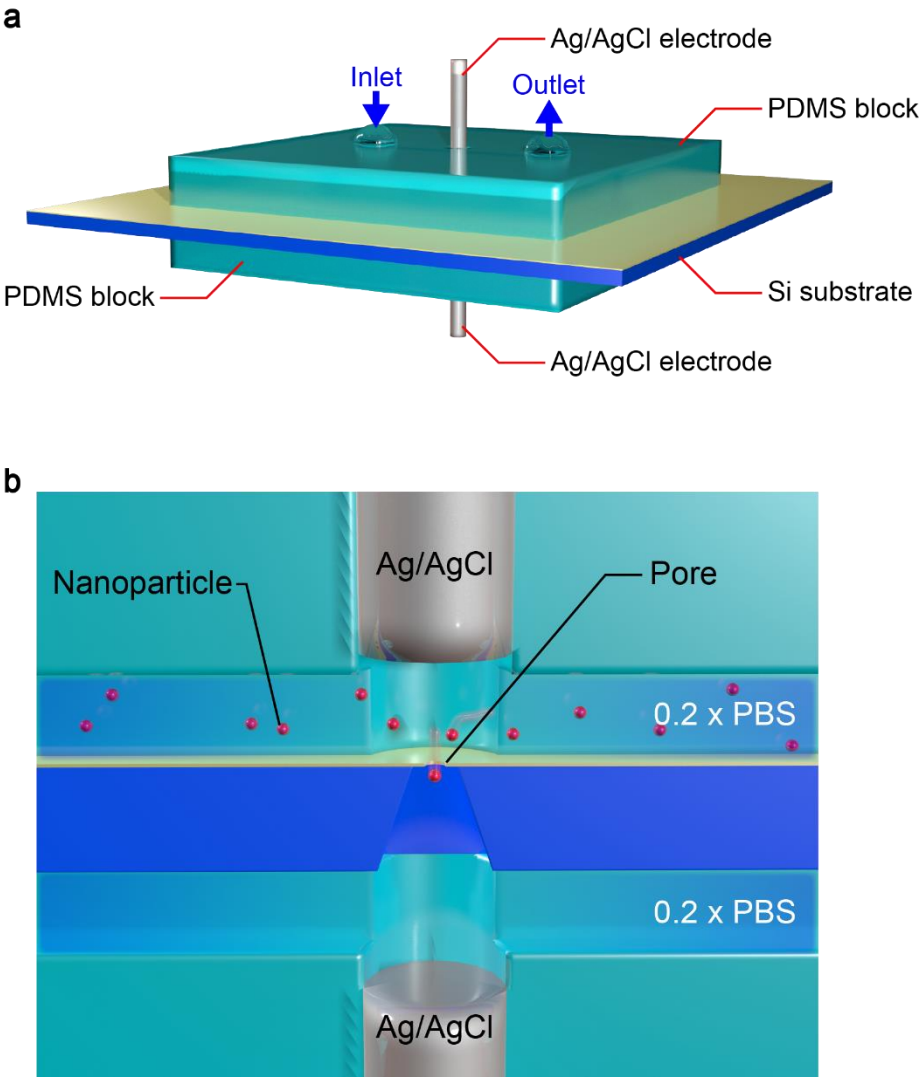

Figure S2. Schematic illustration depicting the measurement set up used for the resistive pulse analyses of single-polymeric nanoparticles using solid-state multipores. Two PDMS blocks were adhered on the both sides of a Si chip wherein multiple pores of diameter 1.2 μm were sculpted in a 50 nm thick Si_3N_4 membrane by nanofabrication techniques such as electron beam lithography and reactive ion etching. In each block, we drilled three holes. One of them was used to place a Ag/AgCl electrode for the cross-membrane ionic current measurements. The other two were utilized as inlet and outlet to inject dilute solution of target particles. During the experiments, one side was filled with 0.2 x PBS containing the test particles while filling the other side with 0.2 x PBS without any particles.

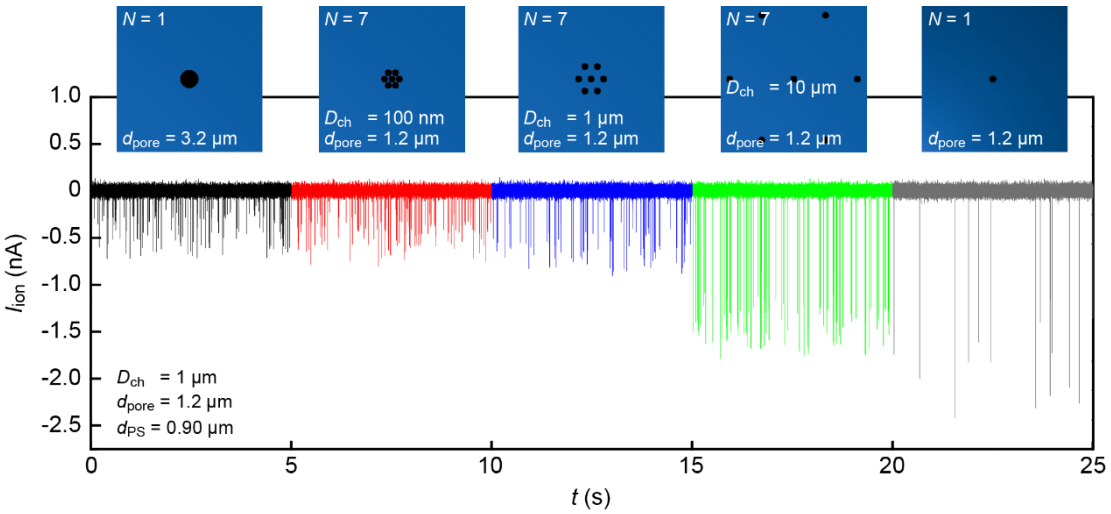

Figure S3. Partial ionic current (I_{ion}) traces obtained for 900 nm-sized carboxylated polystyrene beads in 0.2 x PBS with seven-pores of various inter-channel distance D_{ch}. As the inter-channel distance D_{ch} becomes shorter, the pulse height tends to be weaker and become comparable to those observed in the equivalent-size single-pore (black).

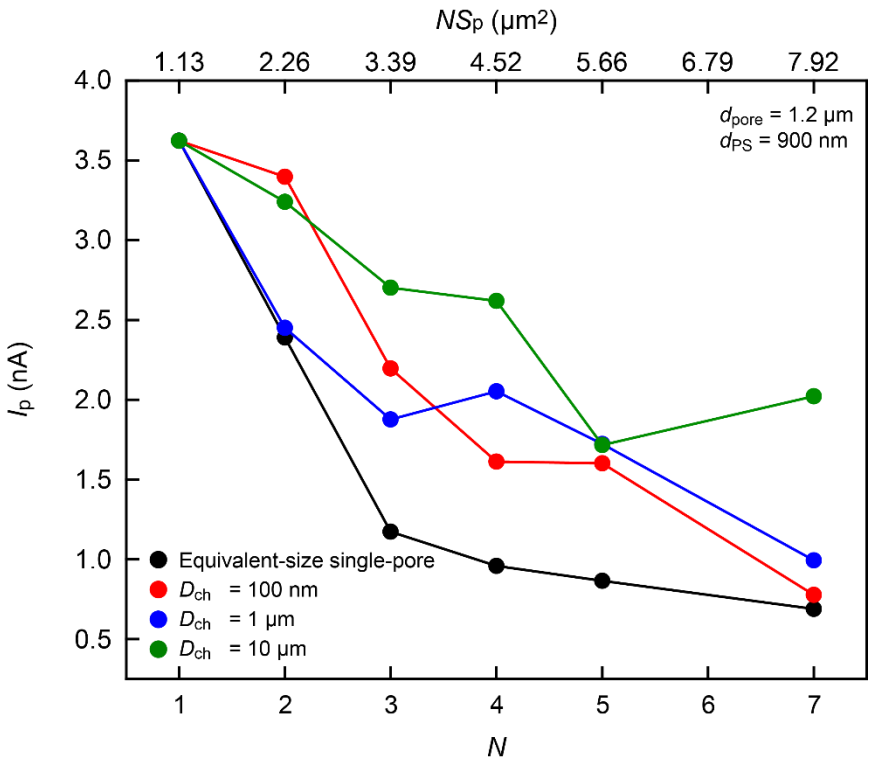

Figure S4. Dependence of the resistive pulse height I_p on the number of channels N with various inter-channel distance D_{ch} conditions for 900 nm-sized carboxylated polystyrene nanobeads in 0.2 x PBS. Data for equivalent-sized single pore of area NS_p are also shown where S_p = 1.13 μm^2 is the cross-sectional area of a 1.2 μm-sized pore. Similar to the case of 780 nm-sized polymeric nanoparticles (Fig. 2c), I_p tends to decrease monotonically with N.

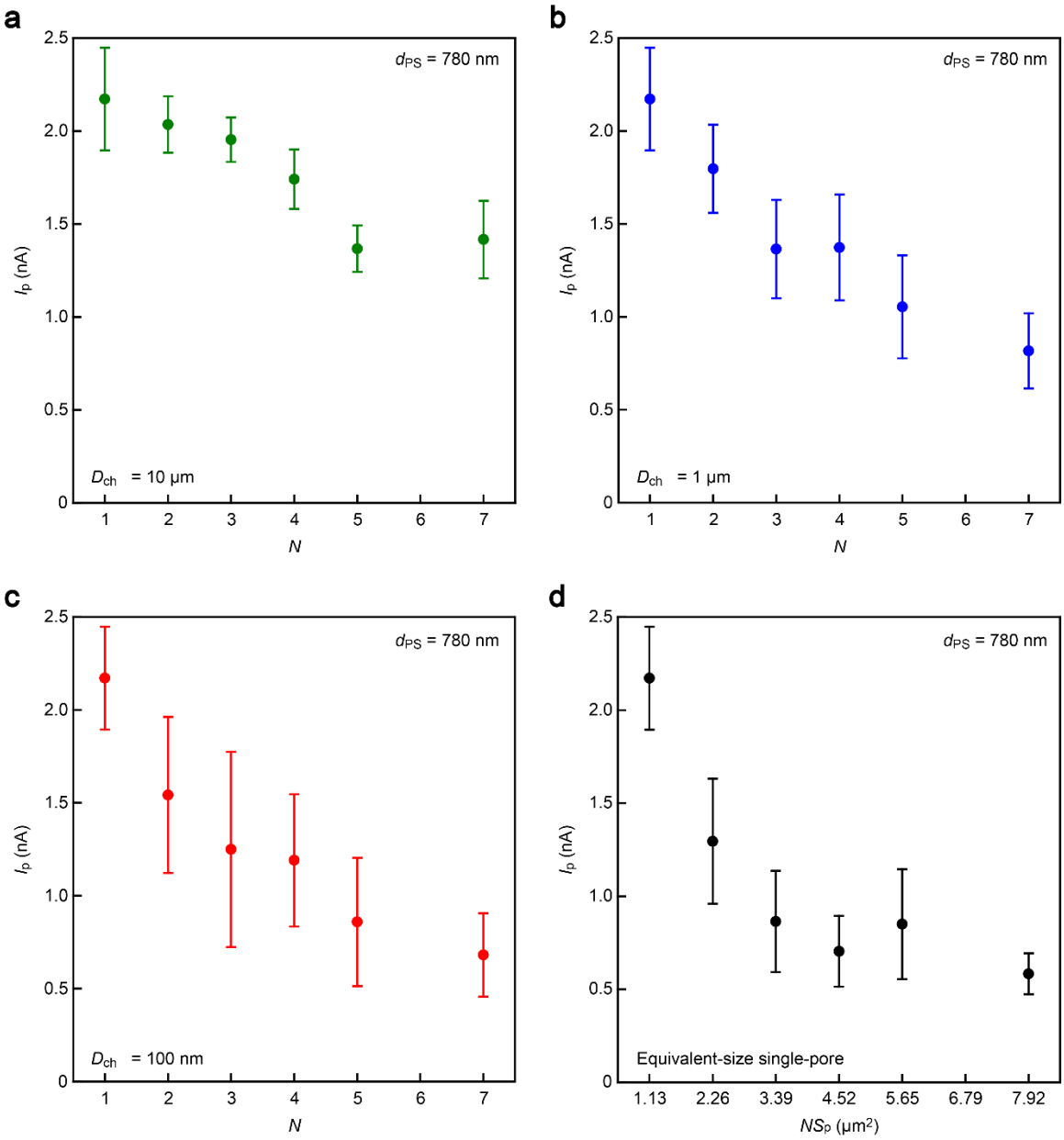

Figure S5. **a-c,** Dependence of the resistive pulse height I_p of carboxylated polystyrene beads of diameter d_{PS} = 780 nm in 0.2 x PBS under 0.1 V on the number of 1.2 μm-sized pores N with inter-channel distance D_{ch} of 10 μm (a), 1 μm (b), and 100 nm (c). **d,** The same plots for the equivalent-sized single pore of area NS_p where S_p = 1.13 μm² is the cross-sectional area of a 1.2 μm-sized pore. Error bards denote the full width at half maximum of Gaussian distributions fit to the I_p histograms.

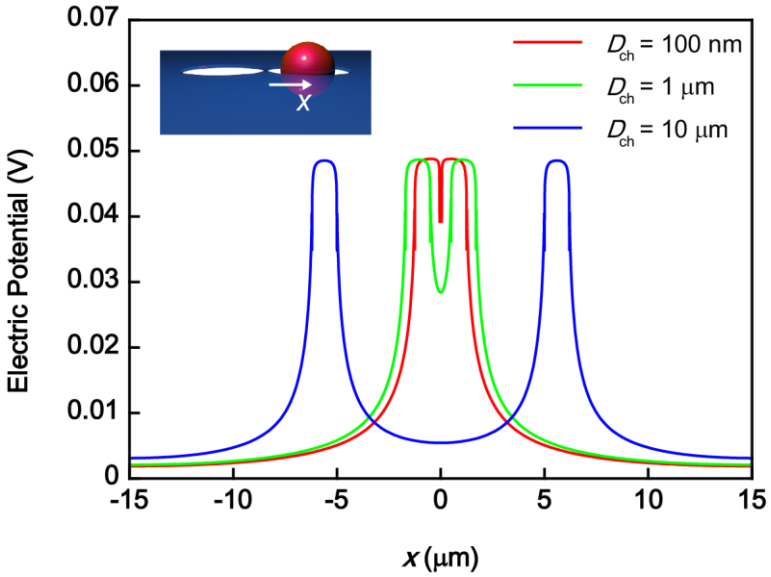

Figure S6. Electric potential distributions in 1.2 μm-sized double-pores of different inter-channel spacing D_{ch} in a 50 nm-thick Si_3N_4 membrane under bias voltage of 0.1 V in 0.2 x PBS simulated using COMSOL. The potential is at the top surface of the membrane. x is along the radial direction centered at the middle of the two channels. Note that the distributions are well-separated when D_{ch} = 10 μm but tend to merge into one peak as the spacing is made shorter.

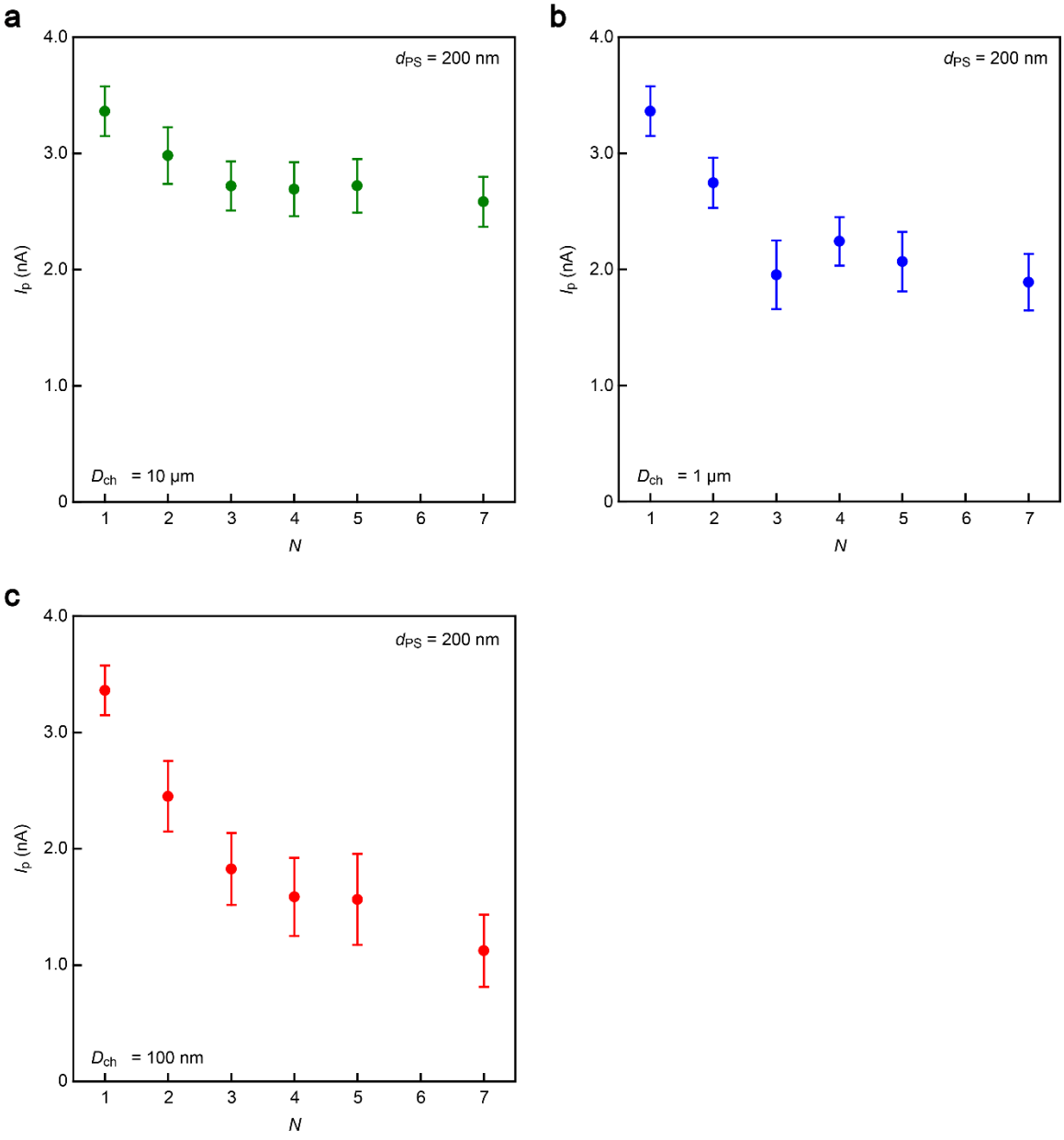

Figure S7. **a-c,** Dependence of the resistive pulse height I_p of carboxylated polystyrene beads of diameter d_{PS} = 200 nm in 1 x PBS under 0.1 V on the number of 300 nm-sized pores N with inter-channel distance D_{ch} of 10 μm (a), 1 μm (b), and 100 nm (c). Error bards denote the full width at half maximum of Gaussian distributions fit to the I_p histograms.

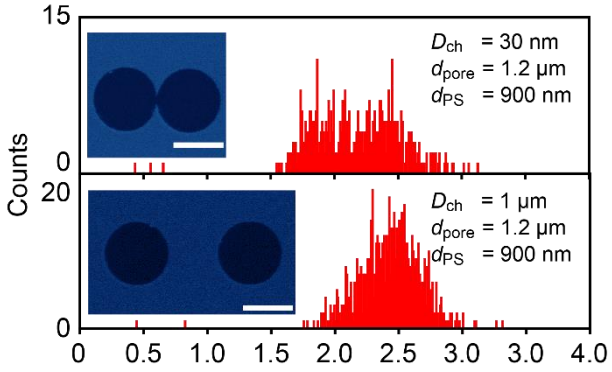

Figure S8. Resistive pulse height (I_p) histograms obtained for carboxylated polystyrene nanoparticles of diameter d_{PS} = 900 nm in 0.2 x PBS with 1.2 μm-sized double-pores of inter-channel distance D_{ch} = 30 nm (top) and 1000 nm (bottom). A bimodal distribution was obtained with D_{ch} = 30 nm while that with D_{ch} = 1 μm demonstrates a single-peak profile. Bin size is 0.01 nA.

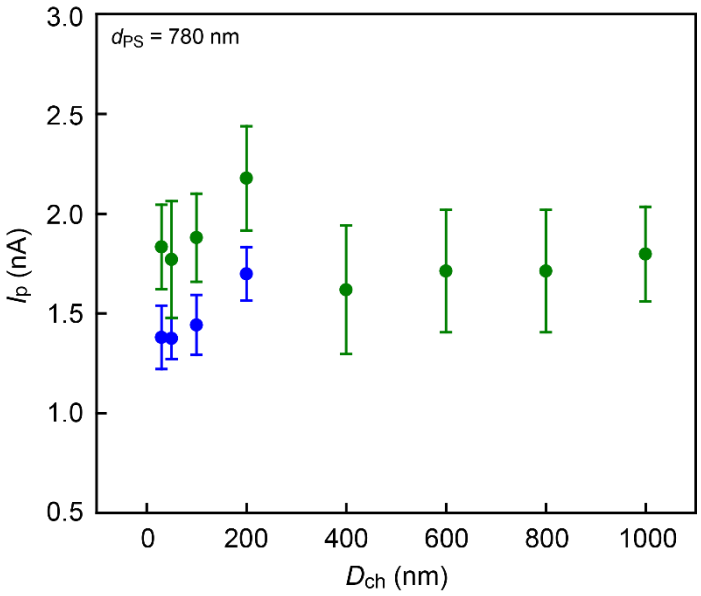

Figure S9. Dependence of the resistive pulse height I_p of 780 nm-sized polystyrene beads in 0.2 x PBS under 0.1 V on the number of 1.2 μm-sized double pores of variable inter-channel distance D_{ch}. Error bards denote the full width at half maximum of Gaussian distributions fit to the I_p histograms.

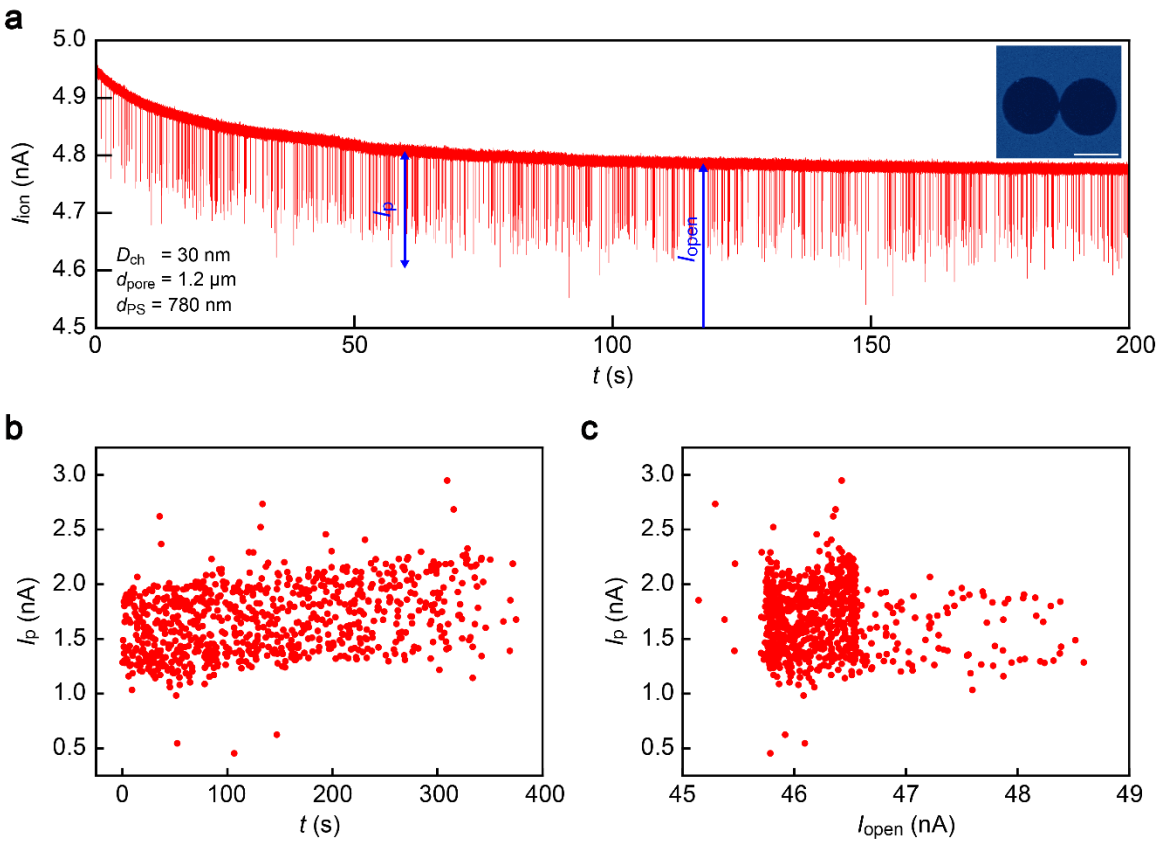

Figure S10. **a,** Ionic current curve obtained for 780 nm-sized carboxylated polystyrene nanoparticles in 0.2 x PBS using a closely-separated (D_{ch} = 30 nm) double-pore. No abrupt drops in the open pore current I_{open}, i.e. the ionic current flowing through the 1.2 μm-sized channels, were observed indicating no channel clogging occurred during the measurement.[S1-S4] **b-c,** The resistive pulse height I_p of each signals shown in (a) plotted as a function of the measurement time t (b) and I_{open} (c). No notable correlation can be found in the plots manifesting the random nature in the emergence of the bimodal I_p states.

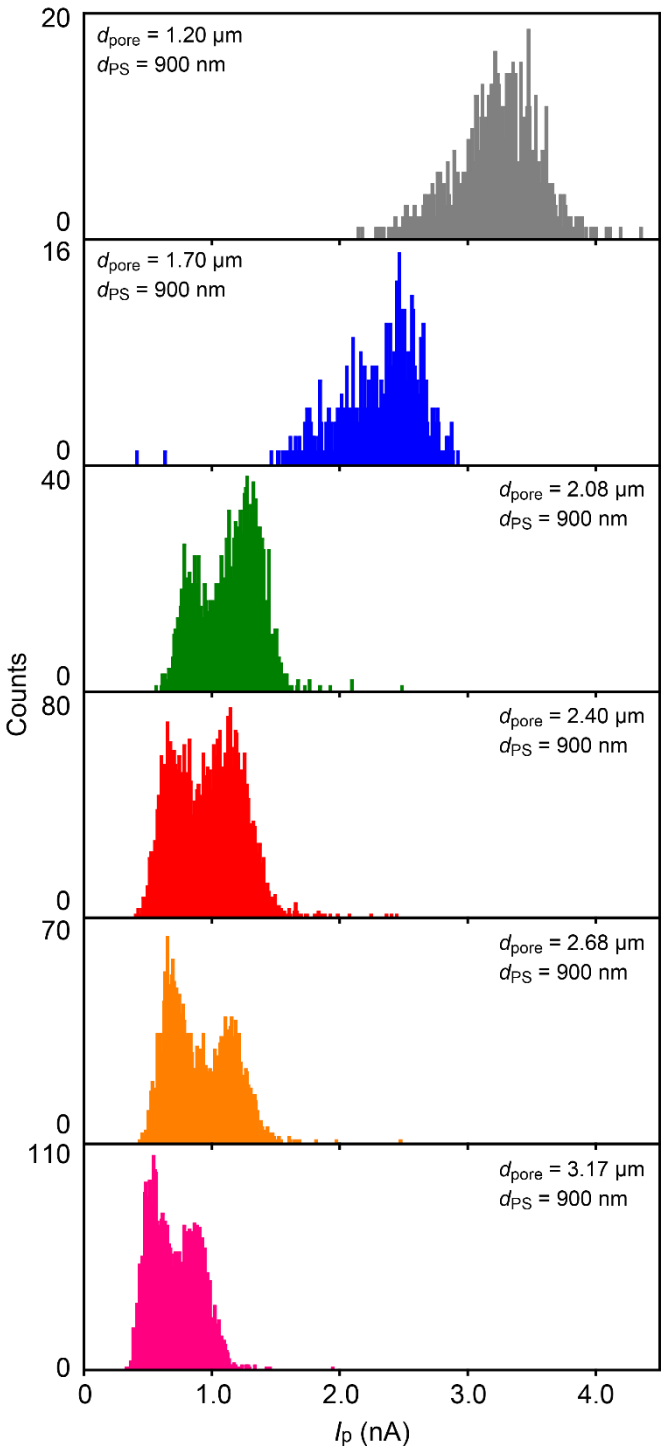

Figure S11. Histograms of the resistive pulse height I_p obtained for 780 nm-sized carboxylated polystyrene beads in 0.2 x PBS under 0.1 V with single pores having equivalent area to those of 1.2 µm-sized one- (grey), two- (blue), three- (green), four- (red), five- (orange), and seven-pores (pink) in a 50 nm thick Si_3N_4 membrane. Note that the distributions start to show additional peak at lower I_p

as the diameter of the equivalent-area pore becomes larger than 1.2 μm. Considering the off-axis effects on the ionic current blockade, it implies that the particles tend to pass through the positions close to the center of the channel more frequently as the pores become larger.

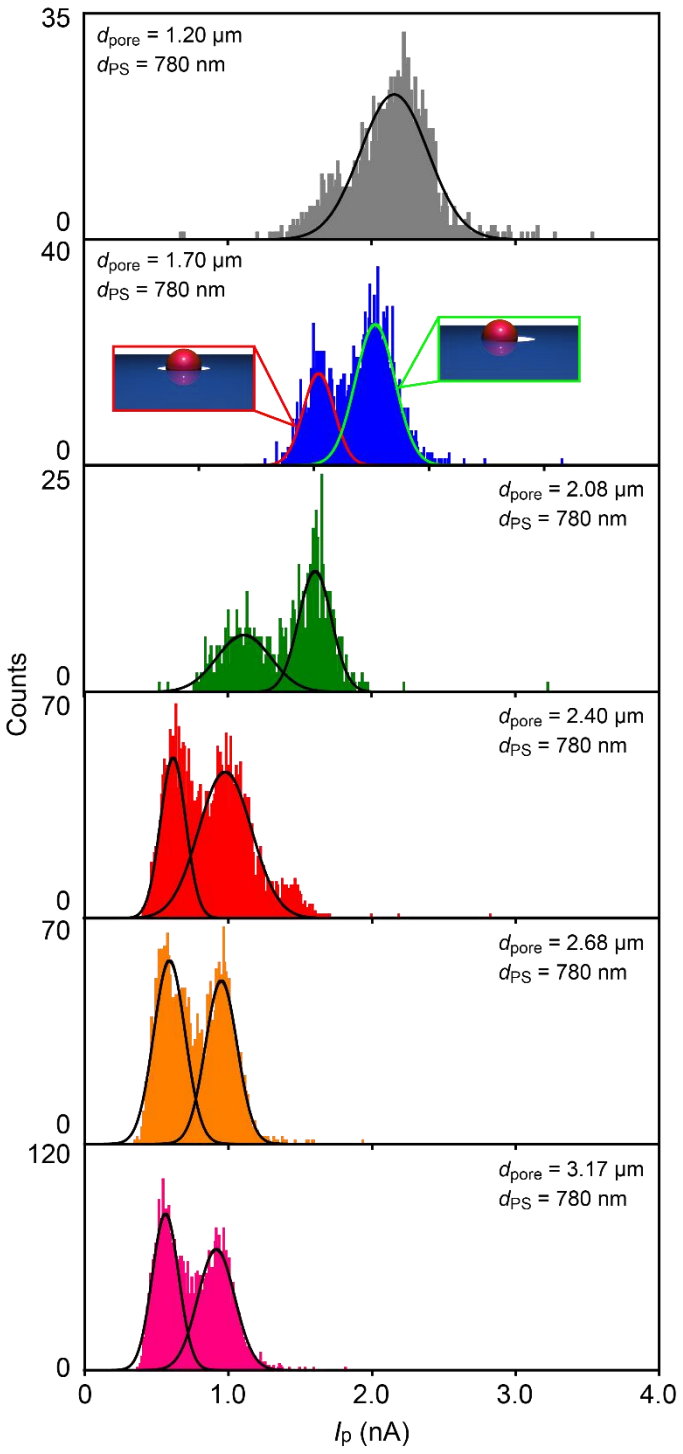

Figure S12. Histograms of the resistive pulse height I_p obtained for 900 nm-sized carboxylated polystyrene beads in 0.2 x PBS under 0.1 V with single pores having equivalent area to those of 1.2 µm-sized one- (grey), two- (blue), three- (green), four- (red), five- (orange), and seven-pores (pink) in a 50 nm thick Si_3N_4

membrane. Note that the distributions start to be bimodal as the diameter of the equivalent-area pore becomes larger than 2.08 μm.

Supplementary references

S1. Kasianovicz, J. J.; Henrickson, S. E.; Weetall, H. H.; Robertson, B. Simultaneous Multianalyte Detection with a Nanometer-Scale Pore. *Anal. Chem.* **2001**, *73*, 2268-2272.

S2. Can den Hout, M.; Krudde, V.; Janssen, X. J. A.; Dekker, N. H. Distinguishable Populations Report on the Interactions of Single DNA Molecules with Solid-State Nanopores. *Biophys. J.* **2010**, *99*, 3840-3848.

S3. Vlassarev, D. M.; Golovchenko, J. A. Trapping DNA Near a Solid-State Nanopore. *Biophys. J.* **2012**, *103*, 352-356.

S4. Tsutsui, M.; He, Y.; Yokota, K.; Arima, A., Hongo, S., Taniguchi, M., Washio, T. & Kawai, T. Trapping and Identifying Single-Nanoparticles Using a Low-Aspect-Ratio Nanopore. *Appl. Phys. Lett.* **2013**, *103*, 013108.