Supplementary Materials for

Edge-of-chaos learning achieved by ion-electron–coupled dynamics in an ion-gating reservoir

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Sections S1 to S5
Figs. S1 to S6

Other Supplementary Material for this manuscript includes the following:

Movie S1
Section S1. The $I_D-V_G$ and $I_G-V_G$ curves for IGRT

Figure S1 shows the $I_D-V_G$ and $I_G-V_G$ curves for IGRT with double sweep for different channel lengths. The application of gate voltage can modulate accumulated charge at the EDL by supplying/removing Li$^+$ ions at the diamond/LSZO interface. As a result, the resistive state of the diamond channel is greatly modulated and the drain current response changes by three to four orders of magnitude as shown in Fig. S1a. While a significant channel length dependence is observed in the $I_D-V_G$ curves, it is absent in the $I_G-V_G$ curves as shown in Fig. S1b. It is because the $I_G$ was measured by using the common gate and the common source electrodes. The $I_D-V_G$ curves showed no huge hysteresis, which is usually found in ion-gating transistors. It is due to low $V_G$ sweep rate (10 mV/s) used in the present study. Li$^+$ ion response is much faster than the $V_G$ sweep rate, thus the $I_D-V_G$ curves basically correspond to the ones in the steady state at each $V_G$ s. That is why the $I_D-V_G$ curves are not accompanied by a huge hysteresis. In the region where $V_G$ is from 0.2 V to 0.5 V, the $I_D$ curves show slight hysteresis, which reflects the slight hysteresis of $I_G$. This is because a contribution from source to drain channel current (suggested to be below nA due to very low carrier density) to $I_D$ is much smaller than that from $I_G$ (about a few nA).
Fig. S1. Electrical characteristics of the IGRT. a, The $I_D-V_G$ and b, $I_G-V_G$ characteristics of the IGRT with different channel lengths.
Section S2. NARMA2 task demonstrated with 2 different input methods by IGR

We performed the NARMA2 task with two input methods, as shown in Fig. S2a. Method 1 is the same procedure as in the second-order nonlinear dynamic equation task, where $u(k)$ is converted to a pulse voltage signal $V_G(k)$ and input to the device (w/o-inversion pulse). Increasing physical nodes is effective for high performance RC. In addition to the normal reservoir states obtained in Method 1, Method 2 uses additional reservoir states, which are obtained by inversion pulses. The procedure is as follows; $u_{inv}(k)$ with the intensity of $u(k)$ inverted as shown in equation (S1) was converted to a pulse voltage signal $V_{inv}(k)$ and input to the IGRT, and an additional reservoir state was obtained by the virtual node method from an additional 8 drain current responses (w/-inversion pulse). To compare these methods without any contribution from differences in network size, the total number of reservoir states for Method 1 and 2 were kept to 80 nodes. However, Fig. 3 shows the results from Method 2 with 10 virtual nodes (160 nodes in total) to obtain the best performance.

$$u_{inv}(k) = 0.5 - u(k)$$

(S1)

The upper panel of Fig. S2b shows the relationship between the IGRT operating condition and the NMSEs (test phase) of the NARMA2 task in Method 1 (wo/-inversion pulse). Good prediction performance was observed in the operation region with an input pulse period of 20 ms or longer and a duty ratio of 25% or higher. In particular, the best prediction performance (NMSE=0.062 test phase) was achieved at a pulse period of 20 ms and a duty ratio of 50%. The target waveform and the predicted waveform of the IGR during the test phase under this condition are shown in the upper panel of Fig. S2c. These two waveforms are in good agreement, indicating that IGR successfully predicted the time series generated by the NARMA2 system. The lower panel in Fig. S2b shows the relationship between the IGRT operating conditions and the NMSEs (test phase) of the NARMA2 task in Method 2 (w/-inversion pulse). Compared to Method 1, the prediction performance improved overall, with good prediction performance in the operation region with an input pulse period of 20 ms or longer and a duty ratio of 75% or higher. In particular, the best prediction performance (NMSE=0.023 in test phase) was achieved at a pulse period of 50 ms and a duty ratio of 75%. The target waveform and the predicted waveform of IGR (test phase) under these conditions are shown in the lower panel of Fig. S2c. Both waveforms are in excellent agreement.
Fig. S2. NARMA2 task demonstrated with 2 different input methods by IGR. a, Schematic illustration of Method 1 (w/o-inversion pulse) and Method 2 (w/-inversion pulse). b, Relationship between IGRT operating conditions and NMSEs in the test phase of the NARMA2 task in Method 1 (upper panel) and Method 2 (lower panel). c, Target and prediction waveforms of the NARMA2 task in Method 1 (upper panel) and Method 2 (lower panel).
Section S3. The effect of device geometry on IGR performance

To evaluate the effect of device geometry on IGR performance, we fabricated an additional device (Device 2) with a different channel geometry than the device described in the main text (Device 1) and evaluated its computational performance. The channel width of Device 2 is twice that of Device 1 (100 µm × 2 = 200 µm) and channel lengths are half that of Device 1 (10 µm, 25 µm, 35 µm, 50 µm, 100 µm, 250 µm, 350 µm, and 500 µm). Optical microscope images of the channel and electrode portions of Device 1 and Device 2 are shown in Fig. S3a. Since the channel resistance of Device 2 is lower than that of Device 1 and channel current is larger in Device 2, there appears no spike in drain current response for all channels as shown in Fig. S3b. As a result, the diversity of reservoir states is reduced, and the best result for Device 2 evaluated in the NARMA2 task worsens to NMSE=0.031 (with inversion-pulse, pulse period: 20ms, duty rate: 50%) compared to the best result for Device 1 (NMSE=0.020) as shown in Fig. 3b. However, the reduced channel resistance increased the operating speed of the IGRT, resulting in improved computational performance in the region of high input pulse frequency (pulse period=5 ms). Results without inversion pulses for input pulse conditions and NMSE for the NARMA2 task on Device 2 are shown in Fig. S3c. It can be seen that device 2 has a much lower error at 5 ms pulse period (200 Hz) compared to device 1 as shown in upper panel of Fig. S2b. This result suggests that IGR can operate over a wide speed range by adjusting the device geometry for required tasks and operation speed.
Fig. S3. The effect of device geometry on IGR performance a, Optical microscope images of Device 1 (left panel) and Device 2 (right panel). b, Various drain current responses of the IGRT at different channel lengths $I_G-V_G$ characteristics of the IGRT (Device2) with different channel lengths. c, The relationship between IGRT (Device2) operating conditions and NMSEs in the test phase of the NARMA2 task in Method1 (without inversion-pulse).
Section S4. Simulation of ion-electron coupled dynamics in IGR

The ion and electron dynamics in our IGR were simulated using the COMSOL multiphysics software application in order to clarify the underlying mechanism in the unique $I$-$V$ characteristics of our device. The EDLT model, which is comprised of Li$^+$ electrolyte, channel and electrodes, was constructed by assuming the physical properties LSZO (Li$^+$ concentration: $10^{22}$ cm$^{-3}$, mobility: $4 \times 10^{-13}$ cm$^2$/V s), EDL (constant capacitance: 4.0 µF/cm$^2$), and the device structure, some of which were modified to reduce computational load. Please refer to the Method section for details of the calculation. After tuning the device parameters, the $I_D$-$V_G$ and $I_D$-$V_D$ characteristics of the simulated model shown in Figs. S4a and S4b agreed well with the experimental result shown for reference.

In the initial state ($t=1.75$ ms indicated as (i) in Fig. S5) of both channel (corresponds to diamond channel of the EDLT) and electrolyte (corresponds to LSZO of the EDLT), significant in-plane carrier distribution was found, in which the densities of positively charged holes and negatively charged Li vacancies are higher near the source electrode than near the drain electrode. This corresponds to the formation of EDL, which is differently charged by voltage distribution due to application of $V_D$ ($=-500$ mV) between the source and drain electrodes. It is noted that any out-of-plane distribution of excess Li$^+$ (and Li vacancy) accumulates within 0.3 nm from the interface. The extremely thin nature of the EDL is consistent with the result of in situ HAXPES observation of LSZO/Au interface [33]. During application of the first $V_G$ pulse [from $t=2$ to $12$ ms, (ii) to (v) in Fig. S5], Li$^+$ moves towards the channel/electrolyte interface, the Li$^+$ accumulated region in the electrolyte and the hole depletion region in the channel gradually proceed from the drain electrode to the source electrode. When the opposite occurs in Li$^+$ density and hole density, $V_G$-driven EDL charging occurs in the vicinity of the channel/electrolyte interface through electrostatic interaction between negatively charged Li vacancies and positively charged holes. After removal of the 1st pulse [from $t=12$ to $22$ ms, (vi) to (x) in Fig. S5], the reverse process takes place in both the electrolyte and the channel. The comparison between (i) the initial state ($t=1.75$ ms) and a long time period after the removal of the 1st pulse (x) ($t=21.75$ ms) in Fig. S5 evidences that the carrier distributions are in two different states, meaning that reverse processes ($12$ ms<$t$<$22$ ms) have longer relaxation times than forward process ($2$ ms<$t$<$12$ ms), which provides good short term memory to store the input history. This is consistent with the drain current response shown in Fig. 4b.
Fig. S4. Simulated electrical characteristics of the EDLT model. a, $I_D-V_G$ and b, $I_D-V_D$ characteristics of the simulated EDLT model. The dotted line shows the experimental result by IGRT.
Fig. S5. The ion and hole distribution at the electrolyte/channel interface during the 1st pulse operation in Fig. 4b.
Section S5. Virtual node dependence in IGR computation performance

To investigate the origin of such high computational performance of the IGR, we evaluated the virtual node dependence of NMSE for the NARMA2 task shown in Fig. S6a. Only physical nodes without inversion pulses were used here to predict the NARMA2 system (8 reservoir states). The blue line in the figure shows the NMSE with all nodes (80 reservoir states). The best prediction performance was obtained for virtual nodes 1 and 6, which correspond to the spike behavior of the drain current, as shown in Fig. S6a. This indicates that such spikes are not just noise, but also contribute significantly to the computation. It has in fact been reported that the effect of noise in RC is to reduce computational performance [60]. As shown in the top panel of Fig. S6b, the reservoir state of virtual node 1, which had the best computational performance, is rich in diversity and nonlinearly transforms the input signal to higher dimensions, while the reservoir state of virtual node 5, which had the worst performance, showed relatively low diversity. The highly efficient computation of IGR is achieved by said virtual and physical nodes, which effectively extract the features of the input signal through the EDL effect.
**Fig. S6. Virtual node dependence in IGR computation performance**

**a,** Virtual node dependence of NMSE in the NARMA2 task. The dotted lines indicate the normalized drain current, done so that the obtained virtual node location corresponds to the NMSE. **b,** Reservoir states obtained from virtual node 1 (upper panel) and 5 (lower panel) correspond to Fig. 2c.