Supplementary information for the manuscript. The experimental set-up and additional parameters of domain chains are described in section I. The mathematical derivation of charge-controlled depolarization fields is given in Section II. Regularization fit is described in Section III. Possible applications of these systems for computing architectures are delineated in Section IV.

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I. Experimental details and data analysis

Polarization reversal under the action of electric field produced by conductive tip of scanning probe microscope (SPM) has been studied in a 20-μm-thick plate of the periodically poled single-crystalline congruent lithium niobate (CLN) with 10 μm period and domain walls strictly oriented along the Y crystallographic direction. The periodical poling has been realized in 0.5 mm thick single-domain Z-cut wafer (Crystal Tech, USA) by conventional poling procedure using a liquid electrolyte (saturated water solution of LiCl)\textsuperscript{1} (Labfer Ltd, Russia). The sample thickness has been reduced to 20 μm by thorough mechanical polishing.

The experiments were carried out in the Center for Nanophase Materials Sciences (Oak Ridge National Laboratory, USA) using commercial scanning probe microscope (Nanoman, Bruker, USA). Piezoresponse force microscopy (PFM) was used for domain visualization. The modulation voltage with amplitude 1 V and frequency ranged from 290 kHz to 350 kHz was applied to SPM tip. Multi-75-G-E SPM tips (Budget Sensors, USA) with conductive platinum coating and nominal radius of tip curvature $R_{\text{tip}} < 25$ nm have been used. Writing of rectangular 2D arrays of the isolated domains was carried out by triangular bipolar pulses with amplitude $U$ ranged from 20V to 100V and duration $t_{\text{sw}} = 250$ ms. Experiments were carried out in ambient conditions (25°C and 40% of humidity), at elevated temperatures up to 150°C and at specified humidity varying from 0 to 90%. The original Bruker thermo table has been used for sample heating. General Electric MG110 hygrometer calibration tool was used in order to produce different humidity levels in the sample cell. To get insight into transient behaviours, the hysteresis loops have simultaneously been measured\textsuperscript{1} in the band excitation PFM mode (not shown).\textsuperscript{2,3}
The distance between neighboring switching points in the line $d$ was ranged from 50 nm to 1 μm, while the distance between lines in the matrix was fixed and equal to 1.25 μm. Measurements in this configuration allowed us to study domain-domain interaction in single line and exclude interaction between domains from different lines. The motion of the grounded SPM tip between switching points has been realized in the contact with surface of the sample.

The sizes of formed domains were found to be strongly dependent on the distance between the neighboring domains (Fig. S1a-c). The formation of uniform domain chains was observed at distances above 50 nm between domain walls of neighboring domains (Fig. S1a). The effective radius, $r$, used for quantitative characterization of the domain size was calculated from domain area $A$ in approximation of round-shaped domain $r = \sqrt{A/\pi}$. The linear dependence of the domain radius in uniform chain on switching pulse amplitude $U$ has been revealed (Fig. S2). These results, obtained for non-interacting domains, are in good agreement with results on switching by the tip explored previously.\(^4\)

![Figure S1](image.png)

**Figure S1.** Domain chains formed after switching with pulse amplitude $U = 50$ V and different distances between switching points at room temperature and 40% humidity. (a) Formation of uniform chain at $d = 500$ nm. (b) Aperiodic chain at $d = 170$ nm. (c) Partial merging at $d = 150$ nm. (d) Linear dependence of domain radius on switching pulse amplitude.
uniform chain; (b) formation of aperiodic domain chain due to domain-domain interaction; (c) partial domain merging. (d) Schematic phase diagram of switching behavior as a function of the voltage and switching point spacing. Description is given in the main text.

Domain-domain interaction was observed at shorter distances between switching points and led to the formation of non-uniform chains (Fig. S1b). Decreasing the distance between switching points below the domain diameter at high pulse amplitudes \((U > 40V)\) led to partial domain merging (Fig. S1c).

![Figure S2](image)

**Figure S2.** Radius of non-interacting domains in the uniform chain vs. switching pulse amplitude. No domains form below \(~20\ V\), the critical threshold for nucleation. Above this threshold, domain size is approximately linear with bias.
These experimental results allow us to plot the schematic phase diagram (Fig. S1d) describing features of the switching behavior in the chain. Application of the pulses with amplitude below a threshold value didn’t lead to domain formation (Region I). Application of the pulses with amplitude above threshold value for relatively long distances between the switching points led to formation of chains with even domain sizes (Region IIIa). Domain-domain interaction at shorter distances between the switching points decreased sizes of some domains in the chain and led to formation of nonuniform domain chains (Region IV). Weak interaction at relatively long distances between the switching points led to formation of the uniform chain of domains with diameter smaller than the first one (Region IIIb). Further decreasing of the distance between the switching points at high pulse amplitudes led to partial domain merging (Region II).

The revealed domain-domain interaction determines the sizes of isolated domains in the chain. Radius of the first domain \( r_0 \) in the chain is determined by the pulse parameters only. The size of the second domain depends both on pulse parameters and on the interaction with the first domain. Thus the size of each domain in the chain correlates with the size of the previous one \( r_{n+1} = q(r_n) \).

Experimental dependences of normalized domain radius \( x_{n+1} \) on normalized radius of the previous one \( x_n \) were presented on the plots (Fig. S3). Normalization was carried out by half of distance between switching points \( d/2: \ x_j = 2r_j/d \). In the case of non-interacting domains (\( U = 40\text{V}; d = 500 \text{nm} \)) experimental points are well localized in the small area in the vicinity of the diagonal \( (x_{n+1} = x_n) \) (Fig. S3a). Domain-domain interaction leads to delocalization of the experimental points (Fig. S3b-e). For alternating domain sizes in the chain (period doubling) the points on the plot are divided between two areas situated symmetrically relatively to the diagonal.
(Fig. S3d). In some cases the diagram has a complex point distribution corresponding to transitions between different modes (period doubling, tripling, etc.) (Fig. S3b,e).

**Figure S3.** Domain chains and plots of the normalized radius of n+1 domain $x_{n+1}$ on normalized radius of previous one $x_n$ for the same pulse amplitude $U = 40$ V and various distances between switching points $d$: (a) 500 nm; (b) 130 nm; (c) 140 nm; (d) 150 nm; (e) 160 nm.

Additional data analysis allows the domain displacement from the switching points to determine whether nucleation proceeds at tip-surface contact to be studied. Displacements of the domains mass centers were calculated relative to the tip position during switching (Fig. S4). Averaged displacement was about 5 nm (Fig. S3d,h). Dependence of the displacement on
domain radius do not show any clear regularity (Fig. S4b,f). This means that observed displacements is caused by non-uniform domain growth due to non-uniformity of the sample surface or by errors of the SPM tip positioning, rather than systematic domain-domain interactions. This in turn allows for Eq. (3) in the main text to be used for description of domain dynamics.

**Figure S4.** (a, e) Images of domain chains; (a, d) domain mass center displacement vs. domain radius; (b, e) domain radius and (c, f) domain mass center displacement vs. the number of domain in the chain. Amplitude of switching pulse $U = 40$ V, distance between domains (a-d) 150 nm; (e-h) 500 nm.
II. Depolarization Field Calculations

II.1. Qualitative analysis of screening process

Initially, the system is characterized by (almost) completely screened polarization. If polarization is close to the surface, polarization charge is positive $+P$ and screening charge is negative, $-(1 - \xi)P$ and $\xi$ is very small ($10^{-5} - 10^{-6}$). The charges are separated by the size of a molecule to yield a dipolar double layer. Overall, the surface is electroneutral or very close to it (deviation from charge neutrality is $\xi P$). During switching, the positive tip provides holes to the surface (and electrons to back electrode, to compensate positive polarization charge on the bottom surface). The tip bias also splits water to produce $H^+$ and $OH^-$, where $H^+$ screens switched and now negative polarization charge. The $OH^-$ are pushed out of the switched domain, as were excess $OH^-$ that were there before, forming a ring around the switched domain. Depending on mobility of the $OH^-$, the lateral transport can be fast or slow compared to the time of the experiment.
(a) **Initial state**  
Vacuum or ultra-dry air  
Screening charges originating from contamination are very sluggish

(b) **Intermediate state 1**  
SPM tip  
Domain formation

(c) **Intermediate state 2**  
Negative screening charges mainly coming from water dissociation on {H\(^+\)+OH\(^-\)} are mobile enough

(d) **Final state**  
Depolarizing field  
NO or very small depolarizing field

(e) **Initial state**  
Ambient air with humidity 30% or more  
Negative screening charges mainly coming from water dissociation on {H\(^+\)+OH\(^-\)} are mobile enough

(f) **Intermediate state 1**  
meniscus  
Domain formation

(g) **Intermediate state 2**  
negative charge excess (ring)

(h) **Final state**  
Depolarizing field

**Figure S5.** Screening charge dynamics during tip-induced switching.
II.2 Electrostatic calculations details

Equation of state \( \mathbf{D} = \varepsilon_0 \varepsilon_g \mathbf{E} \) relates the electrical displacement \( \mathbf{D} \) and electric field \( \mathbf{E} \) in the dielectric gap, \( \varepsilon_g \) is the dielectric layer (gap) permittivity, \( \varepsilon_0 \) is the universal dielectric constant. In a ferroelectric film the electric displacement is \( \mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} \approx \varepsilon_0 \varepsilon_{33}^f \mathbf{E} + \mathbf{P}_s \), where \( \mathbf{P}_s = (0,0,P_3(x,y)) \) is the spontaneous polarization vector. The potential \( \varphi \) of a quasi-stationary electric field, \( \mathbf{E} = -\nabla \varphi \), can be introduced. Inside, the dielectric gap potential \( \varphi \) satisfies Laplace’s equation:

\[
\Delta \varphi_s - \frac{\varphi_s}{R_d^2} = 0, \quad \text{for} \quad -\infty < z < 0, \quad (\text{water electrolyte}) \quad (1a)
\]

\[
\Delta \varphi_s = 0, \quad \text{for} \quad 0 < z < z_0, \quad (\text{dielectric layer}) \quad (1b)
\]

\[
\left( \varepsilon_{33}^f \frac{\partial^2}{\partial z^2} + \varepsilon_{33}^f \Delta \right) \varphi_f = 0, \quad \text{for} \quad z_0 < z < L. \quad (\text{ferroelectric}) \quad (1c)
\]

The Laplace operator is \( \Delta \), \( R_d \) is the Debye screening radius. In Equations (1) we used that \( \operatorname{div} \mathbf{P}_s(x,y) = 0 \) for an uncharged 180-degree domain structure. Equations should be supplemented with boundary conditions of zero potential at \( z << -R_d \) and \( z = L \), \( \varphi_s(x,y,-\infty) \to 0 \) and \( \varphi_f(x,y,L) = 0 \). There are continuous potentials on the boundaries between water and the dielectric layer, \( \varphi_s(x,y,0) = \varphi_s(x,y,0) \), and the dielectric layer and ferroelectric, \( \varphi_s(x,y,z_0) = \varphi_f(x,y,z_0) \). A continuous normal component of displacement is assumed on the boundaries, namely \( -\varepsilon_s \left( \partial \varphi_s / \partial z \right) + \varepsilon_g \left( \partial \varphi_s / \partial z \right) = 0 \) at \( z = 0 \) and \( -\varepsilon_0 \varepsilon_{33}^f \left( \partial \varphi_f / \partial z \right) + P_3 + \varepsilon_0 \varepsilon_g \left( \partial \varphi_s / \partial z \right) = 0 \) at \( z = z_0 \).
Figure S6. Geometry of the problem for uniaxial ferroelectric.

The general solution of Eq.(1) is valid for arbitrary distribution $P_3(x, y)$ is derived as:

$$\phi_s(x, y, z) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \varphi_0(k_x, k_y) \exp\left(ik_x x + ik_y y + z \sqrt{k^2 + \frac{1}{R_d^2}}\right),$$  \hspace{1cm} (2a)

for $z_0 < z < L$:

$$\varphi_g(x, y, z) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \varphi_1(k_x, k_y) \exp(-zk) \exp(\varphi_2(k_x) \exp(\varphi_3(k_y)) \exp(ik_x x + ik_y y),$$  \hspace{1cm} (2b)

$$\varphi_f(x, y, z) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \varphi_3(k_x, k_y) \sinh\left((L-z)\frac{k}{\gamma}\right) \exp(ik_x x + ik_y y)$$  \hspace{1cm} (2c)

Here $\gamma = \sqrt{\varepsilon_{33}/\varepsilon_{11}}$ is the dielectric anisotropy factor and $k = \sqrt{k_x^2 + k_y^2}$. Functions:

$$\varphi_0 = \frac{\varepsilon_s \tilde{P}_s \gamma}{\varepsilon_0 \text{Det}(k)} \sinh\left(\frac{l}{\gamma}\right)$$  \hspace{1cm} (3a)

$$\varphi_1 = \frac{\tilde{P}_s \gamma}{2\varepsilon_0 k \text{Det}(k)} \left(\varepsilon_s k - \varepsilon_s \sqrt{k^2 + \frac{1}{R_d^2}}\right) \sinh\left(\frac{l}{\gamma}\right)$$  \hspace{1cm} (3b)

$$\varphi_2 = \frac{\tilde{P}_s \gamma}{2\varepsilon_0 k \text{Det}(k)} \left(\varepsilon_s k + \varepsilon_s \sqrt{k^2 + \frac{1}{R_d^2}}\right) \sinh\left(\frac{l}{\gamma}\right)$$  \hspace{1cm} (3c)
\[ \varphi_3 = \frac{\vec{P}_3(k)}{\varepsilon_0 k \text{Det}(k)} \left( \varepsilon_g k \cosh(kh) + \varepsilon_s \sqrt{k^2 + \frac{1}{R_d^2}} \sinh(kh) \right) \]  \quad (3d)

\[ \text{Det}(k) = \left\{ \begin{array}{l}
\varepsilon_{33}^f \cosh \left( \frac{k}{\gamma} \right) \left( \varepsilon_g k \cosh(kh) + \varepsilon_s \sqrt{k^2 + \frac{1}{R_d^2}} \sinh(kh) \right) \\
-\varepsilon_s \gamma \left( \varepsilon_s \sqrt{k^2 + \frac{1}{R_d^2}} \cosh(kh) + \varepsilon_g k \sinh(kh) \right) \sinh \left( \frac{k}{\gamma} \right)
\end{array} \right\} \]  \quad (3e)

Hereinafter \( l = L - h \) is the true thickness of ferroelectric film and polarization distribution:

\[ P_3(x, y) = \int_{-\infty}^{\infty} dk_x \int_{-\infty}^{\infty} dk_y \vec{P}_3(k_x, k_y) \exp(ik_xx + ik_yy) \]  \quad (4)

For a single-domain ferroelectric the solution of Eqs.\,(1)-(3) has a relatively simple form:

\[ \varphi_s(z) = \frac{-P_3 l \varepsilon_g l}{\varepsilon_0 \left( \varepsilon_g \varepsilon_s l + \varepsilon_{33}^f \left( \varepsilon_s z_0 + \varepsilon_s R_d \right) \right)} \]  \quad \text{for } -\infty < z < 0, \quad (5a)

\[ \varphi_g(z) = \frac{-P_3 l (R_d \varepsilon_s + \varepsilon_s z)}{\varepsilon_0 \left( \varepsilon_g \varepsilon_s l + \varepsilon_{33}^f \left( \varepsilon_s z_0 + \varepsilon_s R_d \right) \right)} \]  \quad \text{for } 0 < z < h, \quad (5b)

\[ \varphi_f(z) = \frac{P_3 \left( \varepsilon_s h + \varepsilon_s R_d \right) (z - L)}{\varepsilon_0 \left( \varepsilon_g \varepsilon_s l + \varepsilon_{33}^f \left( \varepsilon_s z_0 + \varepsilon_s R_d \right) \right)} \]  \quad \text{for } h < z < L. \quad (5c)

Here \( l = L - h \) is the thickness of the ferroelectric film. In the Debye approximation the space charge density \( \rho_s(z) \) in the water layer and the total charge \( \sigma_s \approx \frac{\varepsilon_0 \varepsilon_s}{R_d^2} \int_0^{-\infty} \varphi_s(z) dz \) is given by the elementary expression:

\[ \sigma_s = \frac{-P_3 \varepsilon_g \varepsilon_s l}{\varepsilon_g \varepsilon_s l + \varepsilon_{33}^f \left( \varepsilon_s z_0 + \varepsilon_s R_d \right)}. \]  \quad (6)

Direct analyses of denominators in expressions (5)-(6) tells us that the effective gap \( H \) can be introduced as

\[ H = z_0 + \frac{\varepsilon_g l}{\varepsilon_s} R_d \]  \quad (7)

For the particular case of a cylindrical domain of radius \( r \) and in the effective gap approximation, the corresponding electric field is:
\[ E_{f3}(\rho, z > H) = \int_{0}^{\infty} dk \frac{J_{0}(kp)}{\epsilon_{0}} \times \left( -\tilde{P}_{s}(k) \tanh(kH) \cosh\left( k(L + H - z)/\gamma_{f} \right) - \tilde{P}_{s}(k) \tanh(kH) \cosh\left( kL/\gamma_{f} \right) \right) \]  

Here we used the following Fourier image of the polarization distribution:
\[ \tilde{P}_{s}(k_{x}, k_{y}) = -P_{s} \delta(k_{x}) \delta(k_{y}) 2\pi + 2P_{s} r J_{1}(kr)/k. \]  
\( \epsilon_{0} \) is the universal dielectric constant, \( \epsilon_{g} \) is the background permittivity of the ferroelectric. The effective layer \( 0 \leq z \leq H \) is a transversely isotropic dielectric with the diagonal permittivity tensor \( \{\epsilon_{11}^{d}, \epsilon_{11}^{f}, \epsilon_{33}^{d}\} \), while at \( z > H \), the permittivity tensor is \( \{\epsilon_{11}^{f}, \epsilon_{11}^{f'}, \epsilon_{33}^{f}\} \), \( \gamma_{f} = \sqrt{\epsilon_{33}^{f}/\epsilon_{11}^{f}} \) is the dielectric anisotropy factor.

The z-component of the electric field of the tip is
\[ E_{z}(r) = \frac{Q}{4\pi \epsilon_{0}} \int_{0}^{\infty} dk J_{0}(kp) \left( 2(\kappa_{b} + \kappa_{d}) \exp\left( -k a - k \frac{z}{\gamma_{d}} \right) + 2(\kappa_{b} - \kappa_{d}) \exp\left( -k a - k \frac{2H - z}{\gamma_{d}} \right) \frac{k}{\gamma_{d}} \right) \]  

\[ \left( \kappa_{b} + \kappa_{d} \right)(\epsilon_{e} + \kappa_{d}) - \left( \kappa_{b} - \kappa_{d} \right)(\epsilon_{e} - \kappa_{d}) \exp\left( -k a - k \frac{2H}{\gamma_{d}} \right) \]  

(9)

at \( 0 \leq z \leq H \)

\[ E_{d}(r) = \frac{Q}{4\pi \epsilon_{0}} \int_{0}^{\infty} dk J_{0}(kp) \left( 4\kappa_{d} \exp\left( -k a - k \frac{H}{\gamma_{d}} \frac{z-H}{\gamma_{f}} \right) \frac{k}{\gamma_{f}} \right) \]  

\[ \left( \kappa_{f} + \kappa_{d} \right)(\epsilon_{e} + \kappa_{d}) - \left( \kappa_{f} - \kappa_{d} \right)(\epsilon_{e} - \kappa_{d}) \exp\left( -k a - k \frac{2H}{\gamma_{f}} \right) \]  

(10)

Here \( J_{0} \) is the Bessel function of zero order, \( a = \epsilon_{s} R_{0}/\kappa \) is the effective charge-surface separation (for a tip size \( R_{0} \)), \( \gamma_{d} = \sqrt{\epsilon_{33}^{d}/\epsilon_{11}^{d}} \) and \( \gamma_{f} = \sqrt{\epsilon_{33}^{f}/\epsilon_{11}^{f}} \) are the dielectric anisotropy factors for two layers, and \( \kappa_{d} = \sqrt{\epsilon_{33}^{d} \epsilon_{11}^{d}} \) and \( \kappa_{f} = \sqrt{\epsilon_{33}^{f} \epsilon_{11}^{f}} \) are the effective dielectric constants.
II.3. Derivation of the recurrent formula for domain sizes

The driving force that determines the radius of a new domain \( r_{n+1} \) at location \( \{d, z\} \) from the centre of some exiting domain \( r_n \) (see Fig. S7) depends on \( r_n \):

\[
E_d(r_n, d, z) \approx -\frac{2P_s}{\varepsilon_0} \int_0^{\infty} dk \frac{J_1(kr_n)J_0(kd)}{\varepsilon_{33}^f \cosh(kL/\gamma) \tanh(kH)} \left( \cosh \left( k \frac{L-z}{\gamma} \right) + \gamma \varepsilon_g \sinh \left( k \frac{L}{\gamma} \right) \right)
\]  

(11)

Here the coordinate \( z \geq 0 \), \( H = z_0 + R_d \varepsilon_g / \varepsilon_S \) is the effective gap thickness, where the screening radius \( R_d \) depends on the humidity level of the surface screening layer. \( L \) is the sample thickness, and \( \gamma \) is the dielectric anisotropy. The ferroelectric dielectric permittivity \( \varepsilon_{33}^f \) may be regarded as equal to that of the gap, \( \varepsilon_g \), as a reasonable estimation. For simplicity we may put \( \varepsilon_{33}^f \approx \varepsilon_g \approx \varepsilon_S \approx \varepsilon \).

**Figure S7.** Schematics of the tip and domain geometry.
Roughly for the considered case \( L \gg z \), the equation (1a) can be identically rewritten as:

\[
E_d (r_n, d, z \geq 0) \approx -\frac{2P_s}{\varepsilon_0 \varepsilon} \int_0^{\infty} d \chi J_1 (\chi) J_0 \left( x - \frac{d}{r_n} \right) \frac{\tanh (xH/r_n)}{\tanh (xH/r_n) + \gamma} 
\]  

Equation (12) is illustrated by see Fig. S8a. The approximate analytical expression was derived at \( \gamma = 1 \):

\[
E_d (r_n, d, z) \approx \frac{P_s}{\varepsilon_0 \varepsilon} \left( f(r_n, d, z) - f (r_n, d, z + 2H) \right),
\]  

\[
f(r, \rho, y) = \int_{\rho}^{\infty} dx \frac{y \text{EllipticE} \left( 4x \rho / \left( y^2 + (x + \rho)^2 \right) \right)}{\pi \left( y^2 + (x - \rho)^2 \right) \left( y^2 + (x + \rho)^2 \right)}.
\]

Equations (13) are illustrated by Fig. S8b.
Figure S8. (a) X-profile of the depolarization field z-component calculated numerically for the cylindrical shaped-domain with radius \( r/H = 1 \) and length \( l/H = 100 \) at \( z = 0 \) (solid curve) and just after the surface \( z = 0.2H \) (dashed curve), (b) Depolarization field from the existing domain, \( E_d(r_n, d \geq r_n, 0) \), calculated from Eq.(13) at \( \gamma = 1 \). Different curves correspond to different ratios \( H/r_n = 0.01 \) (red), 0.05 (magenta), 0.1 (blue), 0.5 (green), 1 (black). (c) The depolarization field from the structure “domain + uniformly negatively charged ring” calculated from Eq.(4) at \( H/r_n = 0.1 \) (red), 1 (black). (d) Depolarization field from the structure “domain + uniformly negatively charged ring” calculated from Eq.(14) at \( H/r_n = 1 \) and different ring thickness \( \delta r_n/r_n = 0.5 \) (red), 1 (magenta), 2 (blue), 3 (green), 5 (black).
For the case when an uncompensated sluggish surface charge with 2D-density $\sigma_s$ in a form of a ring with thickness $\delta r_n$ is adjusting and slowly spreading out from the existing domain, Eq. (13) acquires the form of a superposition:

$$E_d(r_n, d, z) \approx \left[ \frac{\sigma_s - P_s}{\varepsilon_0} \left( f(r_n, d, z) - f(r_n, d, z + 2H) \right) - \frac{\sigma_s}{\varepsilon_0} \left( f(r_n + \delta r_n, d, z) - f(r_n + \delta r_n, d, z + 2H) \right) \right]$$  \hspace{1cm} (14)

The amount of charge captured by the moving domain wall of existing domain is unknown, however it may be reasonable to assume that $\pi \left( r^2_n + \delta r^2_n \right) \sigma_s \approx \pi r^2_n P_s$ and $\delta r_n \approx \alpha r_n$. Equation (14) is illustrated by Fig. S8c,d.

In the general case of a smoothly distributed surface charge $\sigma_s(\rho)$, the depolarization field becomes

$$E_d(r_n, d, z) = \frac{P_s}{\varepsilon_0 \varepsilon} \left( \left[ f(r_n, d, z) - f(r_n, d, z + 2H) \right] \right)$$  \hspace{1cm} (15a)

where

$$f(r, d, z) = \int^{\infty} \int dx \cdot xy E \left( \frac{4\pi d}{\rho^2 + (x+d)^2} \right) \left( \sigma_s(x, r) - P_s \Theta(r-x) \right)$$  \hspace{1cm} (15b)

$E(x)$ is the full Elliptic integral of the second order.

The radius $r_{n+1}(z)$ of the new domain can be estimated from the condition

$$\left< E_d(r_n, d, z) \right> + E_{tip}(r_{n+1}, z) = E_{cr}$$  \hspace{1cm} (16)

The SPM tip field, $E_{tip}(r_{n+1}, z)$, can be approximated as [see II.2]:

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\[ E_{\text{tip}}(r,z) \approx \frac{U(a/\gamma)(a + z/\gamma)}{((a + z/\gamma)^2 + r^2)^{3/2}} \]  

(17)

\[ a = \varepsilon z R_0/\kappa \] is the effective charge-surface separation calculated in the sphere-plane model.\(^4\),\(^5\)

\[ R_0 \] is the tip apex curvature, \( \kappa = \sqrt{\varepsilon_1 E_{33}} \). Using Eqs. (17) along with the condition \( \gamma = 1 \) and

\[ \langle E_d(d, r_n, z) \rangle \approx E_d(\delta, r_n, z), \] Eq.(16) acquires the form:

\[ \frac{(U/a)(1 + z/a)}{((1 + z/a)^2 + (r_{n-1}/a)^2)^{3/2}} = E_{cr} - E_d(d, r_n, H + z) \]  

(18a)

Equation (18a) can be identically rewritten as

\[ \frac{r_{n+1}^2}{a^2} = \left( \frac{(U/a)(1 + z/a)}{E_{cr} - E_d(d, r_n, H + z)} \right)^{2/3} - \left( 1 + \frac{z}{a} \right)^2. \]  

(18b)

At the surface \( (z = 0) \), Equation (18b) allows the solution for the dependence \( x_{n+1} = q(x_n) \):

\[ x_{n+1} \approx \sqrt[2/3]{\frac{u^{2/3}}{(1 - g(\delta, x_n, h))^{2/3}} - 1} \]  

(19a)

where

\[ u = \frac{U}{E_{cr}a}, \quad h = \frac{H}{a}, \quad \delta = \frac{d}{a}, \quad x_{n+1} = \frac{r_{n+1}}{a}, \quad x_n = \frac{r_n}{a}. \]  

(19b)

The function \( g(\delta, x_n, h) = E_d(d, r_n, H)/E_{cr} \). The Tailor series of the depolarization field \( E_d(x_n, d, z) \) in Eq.(15) is proportional to the even powers of \( x_n^2 \) from the problem symmetry. Also it is evident that the depolarization field is absent at \( x_n = 0 \) (domain that causes the field is absent) and vanishes with increasing of the relative distance \( \delta \) between the domains as well as in
the case of ideal screening at $H=0$ ($h=0$). Eventually the series terms appeared proportional to the product $(h/\delta)x_n^2$ as:

$$g = \frac{h}{\delta} x_n^2 \left( 1 - \sum_{m=1}^{\infty} \beta_m x_n^{2m} \right) \approx \frac{h}{\delta} x_n^2 \left( 1 - \beta x_n^2 \right)$$

(20)

where $\lambda = \frac{P_s}{\varepsilon_0 E_{cr}}$ is parameter determined by the spontaneous polarization and critical field for the wall motion. Approximate equality in Eq.(20) cuts the series on the second term and so $\chi$ and $\beta$ are expansion coefficients determined by the evident form of the depolarization field function $f(x_n)$ in Eqs.(15b). Dimensionless variables are listed in the Table S1.

Dependences $x_{n+1}(x_n)$ calculated from Eq.(19) for different values of $\mu$ are shown in Figs. S9-S11. Note, that the domain merging in a chain starts at $\delta \leq x_n + x_{n+1}$. So, the condition $\delta = x_n + x_{n+1}$ should be accounted in the iteration procedure based on Eq.(19).

Nucleation of a single domain in the absence (or weak influence) of the pre-existing domain (i.e. at $x_n = 0$ or $\delta \to \infty$) is possible if the normalized bias is more than the critical $\mu > 1$, as the single domain shape is $x_{n+1} = \sqrt{u^{2/3}} - 1$. Note that the law $x_{n+1}(0) \propto \sqrt{u^{2/3}} - 1$ is in agreement with Landauer-Molotskii theory.\textsuperscript{6, 7}

We further explore at which critical bias, $u_m$, the domain merging starts at a given domain separation $\delta$. The corresponding merging boundary $u_m(\delta)$ can be estimated from Eq.(19a) as

$$x_1 = \delta - x_0 \approx \sqrt{\frac{(u_m)^{2/3}}{(1 - g(\delta, x_0, h))^{2/3}}} - 1, \quad x_0 = \sqrt{(u_m)^{2/3}} - 1$$

(21)

The solution of Eq.(21) can be found numerically at fixed $h$ and $\lambda$. 

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Figure S9. Dependence $x_{n+1}(x_n)$ calculated from Eq.(19) for parameters $\delta = 4$, $h = 1$ (a,b) and $h = 0.1$ (c,d), $\lambda = 10$ and different values of parameter $\mu = \mu^{2/3}$ indicated near the curves. Plot (a,c) $\sigma_s = 0$ (no charged ring). The uniformly negatively charged ring exists in plots (b,d) calculated for $\sigma_s \approx \frac{P_s}{(1+\alpha)^2 - 1}$ and $\alpha = \delta r_n / r_n = 2$. The dashed line is the merging line $x_{n+1} = \delta - x_n$. 
Figure S10. (a) Surface charge distribution with $\sigma_s(d, r) = -\frac{r^2(d - r)^2 \theta(d - r) \exp\left(\frac{d - r}{R_{\text{max}}}\right)}{4R_{\text{max}}^3 \left(r + 3R_{\text{max}}\right)}$. (b-c) Depolarization field distribution calculated from using Eq.(15) for parameters $R_{\text{max}} = 0.25r_n$. (d-f) Dependence $x_{n+1}(x_n)$ calculated from Eq.(19) using Eq.(15) for and parameters $\delta = 4$, $\lambda = 10$, $R_{\text{max}} = 0.25a$ and humidity $h = 10$ (d), $h = 10$ (e) and $h = 0.1$ (f), and different values of parameter $\mu = u^{2/3}$ indicated near the curves.
Figure S11. (a) Surface charge distribution with $\sigma_s(\rho, r) = -\frac{\theta(\rho - r)\theta(R_{\text{max}} - \rho)r \exp\left(\frac{(\rho - r)}{r}\right)}{2(2r - \exp\left(1 - \frac{R_{\text{max}}}{r}(r + R_{\text{max}})\right))}$. 

(b-c) Depolarization field distribution calculated from using Eq.(5) for parameters $R_{\text{max}} = 4r$. (d-f) Dependence $x_{n+1}(x_n)$ calculated from Eq.(8) using Eq.(5) for parameters $\delta = 4$, $R_{\text{max}} = 4a$, $\lambda = 10$, and humidity $h = 10$ (d), $h = 10$ (e) and $h = 0.1$ (f), and different values of parameter $\mu = u^{3/2}$ indicated near the curves.
Eq.(19) can be approximated as $x_{n+1} \approx \frac{u^{2/3}}{1 + \frac{h}{\delta} \chi \lambda^2 (1 - \beta \chi^2)} - 1$. Then let us introduce the rescaling in the equation:

$$z_n = c \beta x_n^2 + \frac{1-c}{2}$$

(22a)

where parameter $c = \left( 1 + \sqrt{(w-1)^2 + 4w(m-1)} \right) / w$, parameters $w$ and $m$ are defined as $m = \left( u^{2/3} - 1 \right) \beta + 1$ and $w = 2u^{2/3} (\lambda \chi h / 3 \delta)$. It is worth to note that $w = ml$, where the parameter

$$l = \frac{2u^{2/3} (\lambda \chi h / 3 \delta)}{(u^{2/3} - 1) \beta + 1}.$$  The rescaling (22a) finally yields a well-known logistic map:

$$z_{n+1} = \alpha z_n (1 - z_n)$$

(22b)

Hence, the behaviour of the system is controlled by a single master parameter $\alpha = 1 + \sqrt{(w-1)^2 + 4w(m-1)}$. The properties of the mapping (Eqn. 22b) and corresponding bifurcation diagram are well-studied. For $\alpha$ between 0 and 1, the domain population will eventually die, independent of the initial domain. With $\alpha$ between 1 and 3, the population will approach the value $1 - \alpha^{-1}$. At $\alpha = 3$ there is an onset of period doubling bifurcation, with the onset of quadrupling at $1 + \sqrt{6}$. The subsequent increase of $\alpha$ opens a period doubling cascade. At $\alpha$ approximately 3.57 there is the onset of chaotic behavior, with isolated ranges of $\alpha$ that show non-chaotic stability windows. Beyond $\alpha=4$, the values eventually leave the interval $[0,1]$ and diverge for almost all initial values. By contrast, the solution when $\alpha=2$ is

$$z_n = \frac{1}{2} \left( 1 - 2 \xi_0 \right)^2^z$$

where $0 < \xi_0 < 1$. ☋
Figs. S13a,b is the diagram of the chaotic behaviour of Eq.(22b) in coordinates \( \{m,l\} \). A bifurcation diagram Fig. S12c summarizes this chaotic behaviour of \( z_n(\alpha) \). The horizontal axis shows the values of the parameter \( \alpha \) while the vertical axis shows the possible long-term values of \( z \). Phase diagram of switching behaviour as a function of dimensionless voltage \( u = (U/E_r)a \) and domain spacing \( \delta = d/a \) is shown in Fig. S13d. Note that for small \( \delta \) the domain sizes grows with \( \delta \), and then becomes constant, corresponding to zero interaction.

![Phase diagram](image1)

![Bifurcation diagram](image2)

![Master parameter diagram](image3)

**Figure S12.** (a) Phase diagram of switching behaviour as a function of parameters \( m \) and \( l \). (b) Number of solutions dependent upon parameters \( m \) and \( l \). (c) Bifurcation map as a function of the master parameter \( \alpha \).
**Fitting to experimental diagram Figure 2j.** Using the probe size $R_0 = 30$ nm, obtained as the average from experimental margins 15 – 45 nm (e.g. as determined from the ferroelectric wall width), this scaling allows the critical field be estimated as $E_{cr} = 0.67$ V/nm from the theoretical condition $u_{cr} = \left( \frac{U_{cr}}{E_{cr}R_0} \right) = 1$ (Fig.5c) and experimental value of the critical voltage $U_{cr} = 20$ V (Fig.2j). The value of the critical field is well within margins of a Peierls field calculated as 0.4 – 0.8 V/nm using the Peierls barrier of the domain wall motion $W_p$ of value 125 mJ/m$^2$ calculated by DFT$^{10}$ and 225 mJ/m$^2$ from empirical study (we used that $W_p \propto P_s^2 E_{cr} A_0$, spontaneous polarization $P_s = 0.75$ C/m$^2$, wall width $a_0 = 0.4$ nm). Using the parameter $h=2$, the effective water layer thickness appeared $H = hR_0 = 60$ nm. Dimensionless variables and their best fitting values are listed in the Table S1.

**Table S1.** Dimensionless variables and their best fitting values

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Definition via dimension physical values</th>
<th>Physical sense and the best</th>
<th>Fitting value for figure 5c</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\delta$</td>
<td>$d/a$</td>
<td>ratio of the distance $d$ between the domain centres to probe size $a$</td>
<td>$\delta=2-12$ (y-axes) $d=50-400$ nm (y-axes) $a=30$ nm</td>
</tr>
<tr>
<td>$u$</td>
<td>$U/E_{cr}a$</td>
<td>ratio of the voltage $U$ applied to the probe to the critical voltage $aE_{cr}$</td>
<td>$u=0.5$ (x-axes) $U=20-100$ V (x-axes) $u_{cr} = \left( \frac{U_{cr}}{E_{cr}R_0} \right) = 1$, $U_{cr} = 20$ V, $E_{cr}a = 20$ V, $E_{cr} = 0.67$ V/nm</td>
</tr>
<tr>
<td>$h$</td>
<td>$H/a$</td>
<td>Ratio of the water layer effective thickness $H$ to the probe size $a$. $H$ is defined by the effective humidity</td>
<td>$h=2$, $H=60$ nm</td>
</tr>
<tr>
<td>$x_n$</td>
<td>$r_n/a$</td>
<td>domain radius $r_n$ normalized on the probe</td>
<td>variable</td>
</tr>
</tbody>
</table>

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<table>
<thead>
<tr>
<th>( \lambda )</th>
<th>( \frac{P_S}{\varepsilon_0 E_{cr}} )</th>
<th>size ( a )</th>
<th>( \lambda \approx 3, P_S=0.75 \text{ C/m}^2, \varepsilon=30, \varepsilon_0=8.85 \times 10^{-12} \text{ F/m}^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( g )</td>
<td>( \frac{E_d}{E_{cr}} )</td>
<td>Ratio of the depolarization field to the critical field ( E_{cr} ) that series over ( x_n^2 ), where, ( \chi ) and ( \beta ) are expansion coefficients</td>
<td>Function with parameters ( \beta=0.8, \chi=1 )</td>
</tr>
<tr>
<td>( z_n )</td>
<td>( c \beta x_n^2 + \frac{1-c}{2} )</td>
<td>Rescaling of ( x_n ) to reach a logistic map ( z_{n+1} = \alpha z_n (1-z_n) )</td>
<td>variable</td>
</tr>
<tr>
<td>( \alpha )</td>
<td>( \alpha = 1 + \sqrt{(w-1)^2 + 4w(m-1)} )</td>
<td>Master parameter</td>
<td>Actual range of bifurcations and chaos is 3 - 4</td>
</tr>
<tr>
<td>( c )</td>
<td>( \frac{1+\sqrt{(w-1)^2 + 4w(m-1)}}{w} )</td>
<td>Auxiliary functions of normalized voltage ( u ) and expansion coefficients ( \chi ) and ( \beta )</td>
<td>Parameters ( c, m ) and ( w ) are voltage-dependent, namely both are proportional to ( u^{2/3} )</td>
</tr>
<tr>
<td>( m )</td>
<td>( \left( u^{2/3} - 1 \right) \beta + 1 )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( w )</td>
<td>( 2u^{2/3} \left( \lambda \chi h / 3\delta \right) )</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

III. Regularization smoothing

To smoothen the \( r_{n+1} = q(r_n) \) dependence, we utilize the regularization approach. The function \( \rho(r_n) \) is defined on the grid corresponding to original \( r_n \) values. At each iteration, the element value is changed by \( \delta \rho \) and an error function is calculated. The change is accepted if error is decreased and discarded otherwise.

The error function, EF, is chosen to satisfy two conditions, namely to minimize the least square error between the experimental, \( PR_{\exp} \), and fitted, \( PR_f \) data, and to ensure the smoothness of the resulting data (regularization condition). The EF is represented as:
\[ EF = E_{\text{inst}} + E_\lambda = (r_{n+1} - \rho(r_n))^2 + \lambda D[\rho(r_n)]^2, \]  

where \( D[\rho(r_n)] \) is the derivative of the function and \( \lambda \) is the regularization parameter. It was found that small values of \( \lambda \) allow better fits to the experimental hysteresis loops, but at the expense of a \( \rho(r_n) \) showing multiple non-physical maxima and minima (similar to the high noise values inevitable in direct differentiation). Large values of \( \lambda \) favor smooth \( \rho(r_n) \), but result in large deviations between the fitted and experimental loops.

**IV. Bitwise logic operations**

The period doubling regime shown in Fig. 2(f) of the main text can be used as the basis for binary logic operations. Let us use the convention that the small radius domains correspond to logic 0 and the large radius domains to logic 1. Fig. 2(f) demonstrates that under appropriate conditions (such as the spacing between domains and voltage pulse amplitude) the small domains (logic 0) alternate with large ones (logic 1). Consequently, a domain created in the vicinity of a small domain (logic 0) will be large (logic 1) and vice-versa. It is evident that such functionality corresponds to the NOT gate.

In order to apply the NOT gate to a few bit number \( A \), (i) the binary representation of this number is written in the form of domains in the horizontal direction with a large spacing between adjusting bits (e.g., >500nm), and (ii) the second line of domains (using a constant pulse amplitude) is created close to the first line at the distance corresponding to the period doubling regime (~150nm). According to the period doubling behaviour, 1-s in the second line will corresponds to 0-s in the first line and 0-s in the second line will correspond to 1-s in the first. Therefore, the second line is the bitwise NOT of the input \( A \), namely, NOT(\( A \)). See Fig. S14 (a) for a specific example.
Figure S14. Bitwise logic operations. (a), (b), (c) Examples of bitwise NOT, NOR and AND, respectively. Note that the pulses of lower amplitude are used solely to set logic zeros in the inputs A and B.

The bitwise NOR is realized when 2 input numbers, A and B, are written as two lines at a double period of period doubling regime (~300nm or close to this distance), and then the 3-rd line is created (using a constant pulse amplitude) between these two lines (Fig. S14 (b)). According to the period doubling regime pattern (Fig. 2(f)), 0-s in the central line will correspond to (0,1), (1,0) and (1,1) combinations of corresponding bits in A and B, while 1-s are induced for (0,0) combination only. It is apparent that the central line represents NOR(A,B). Realization of the AND gate requires more steps. Technically, the AND gate is created using the identity AND(A,B)=NOR(NOT(A), NOT(B)).
References