## Supporting Information:

## Towards the limit of ferroelectric nanostructures: switchable sub-10 nm nanoisland arrays

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## 1. BE-PFM images of three different PTO nanoisland arrays



**Figure S1.** (left-hand) Topography, (middle) BE-PFM amplitude, and (right-hand) BE-PFM phase images (size:  $200 \times 200 \text{ nm}^2$ ) of three different PTO nanoisland arrays. The top, middle, and bottom panels are taken from samples *A*, *B* and *C*, respectively.

## 2. Piezoresponse of PTO nanoislands with larger $V_{\rm NI}$

Figure S2 gives topography and BE-PFM phase images for another ultrahigh density array of the PTO nanoislands with  $V_{\rm NI} = 2300 \text{ nm}^3$  ( $d_{\rm NI} = 20.5 \pm 4.0 \text{ nm}$ , and  $h_{\rm NI} = 7.0 \pm 1.0 \text{ nm}$ ). The sample was thermally treated at 500 °C for 15 min to crystallize PTO nanoislands epitaxially on STO:Nb(100) substrate. Heating/cooling rates of 1 °C/s were employed. While only one polarization state was observed in smaller nanoislands, two distinguishable polarization states can be observed in larger nanoislands as shown in Fig. S2(b). Furthermore, it is clearly seen in Fig. S2(c) that a distinct piezoresponse hysteresis loop is observed for a single PTO nanoisland. The results clearly show apparent ferroelectricity of the PTO nanoislands.



Figure S2. (a) Topography and (b) BE-PFM phase images for ultrahigh density array of PTO

nanoislands with  $V_{\rm NI} = 2300 \text{ nm}^3$ . (c) Piezoresponse hysteresis loop for a single PTO nanoisland measured at the red dotted circle in (a). The loop was averaged over 8 measurements.