

1 **SUPPLEMENTARY INFORMATION**

2 Yimeng Li *et al.*

3 **Unveiling Electric-Field-Driven Deformation Dynamics in Metal Nanostructures**

4

5 This supplementary information includes:

6 Supplementary Figure 1-23

7 Supplementary Note 1-6

8 Supplementary Reference

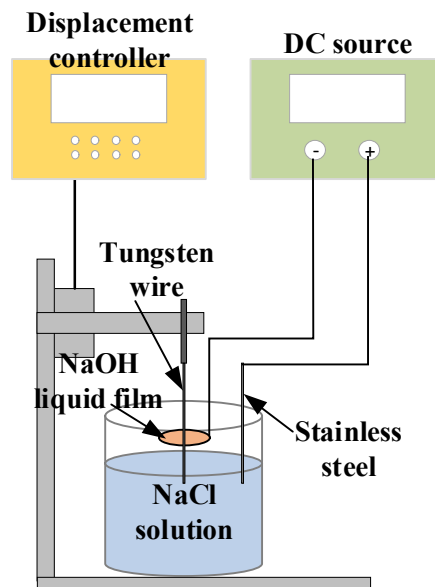
9 Supplementary movie 1

10

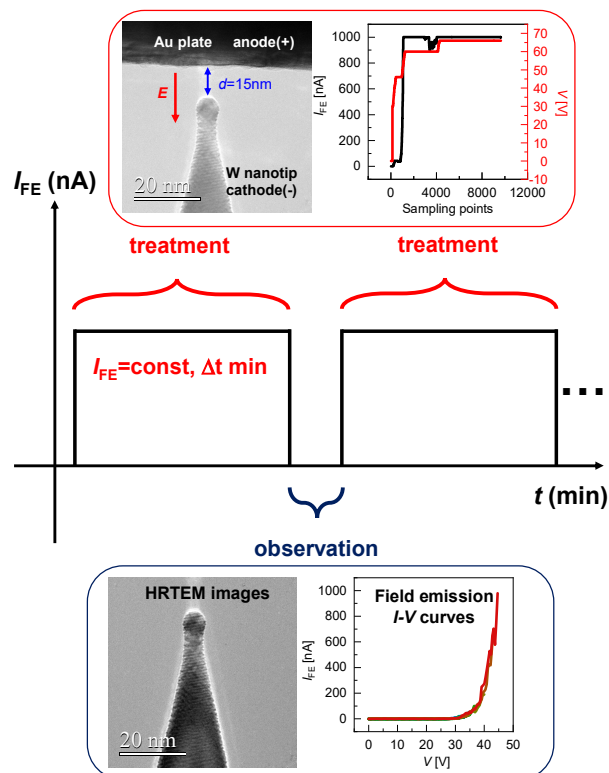
11

12

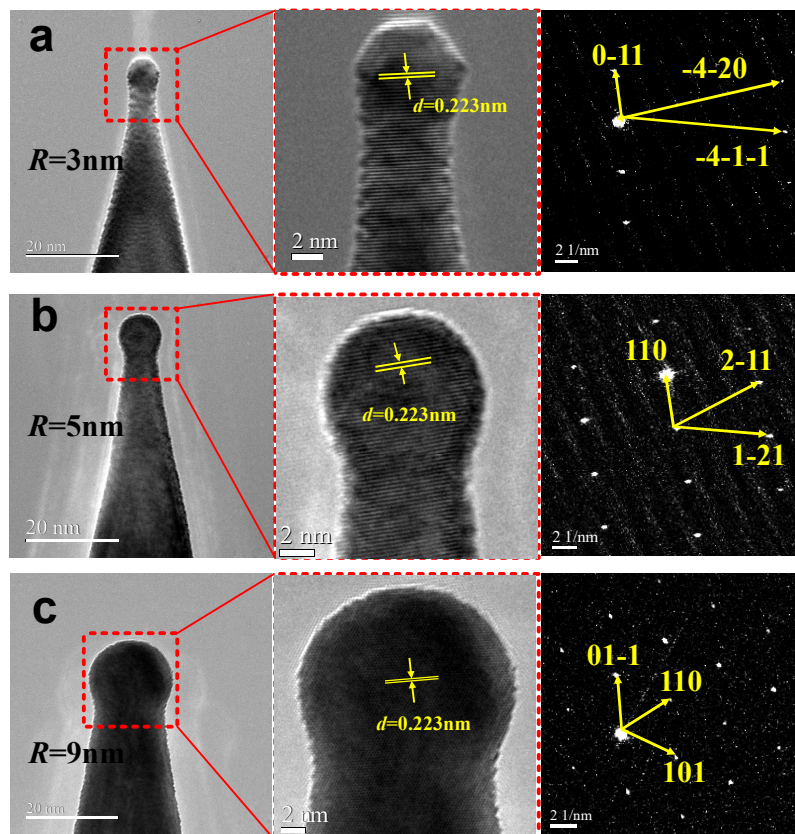
13



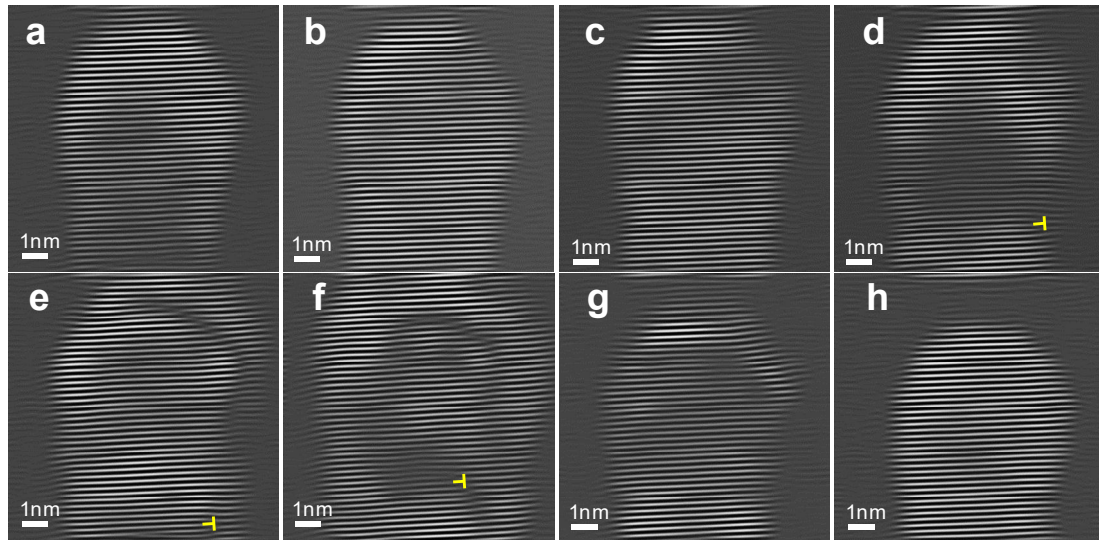
Supplementary Fig. 1| The schematic diagram of double-electrolyte electrochemical etching method.
A pure tungsten wire passes through a NaOH lamella into saturated NaCl, with etching driven by a DC source at the NaOH lamella.



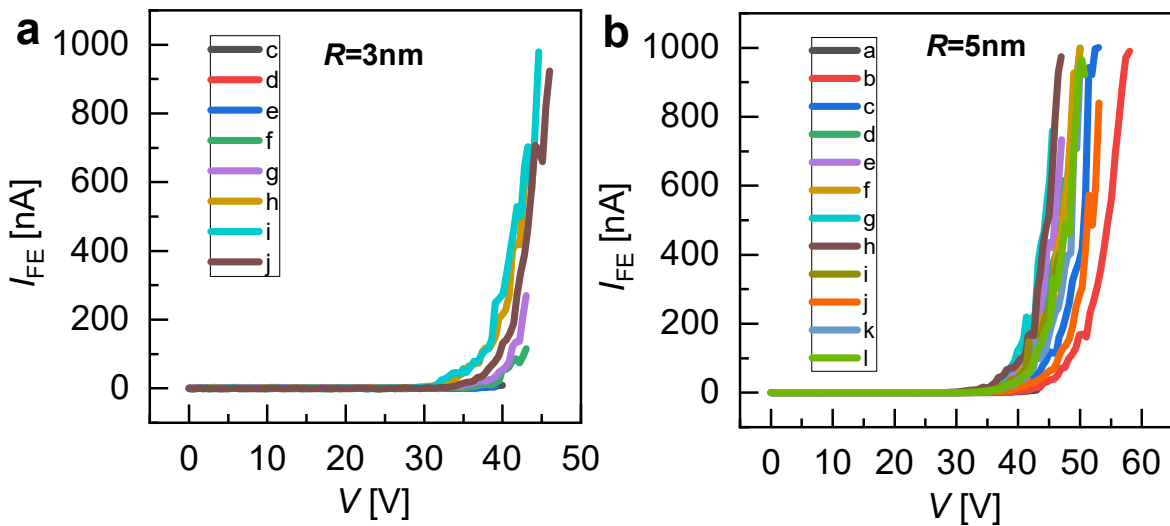
Supplementary Fig. 2| The experimental process for tracking deformation behaviors. The morphology evolution experiment of pure tungsten nanotips is composed of two periods, the treatment period and the observation period. The treatment period maintains a constant field emission current to induce morphology changes. During the observation period, TEM imaging and field emission measurements characterize the evolved nanotip.



Supplementary Fig. 3| The TEM images of the initial tungsten nanotips with different apex radii ($R = 3 \text{ nm}$, 5 nm and 9 nm) and the corresponding post-processed selected area electron diffraction (SAED) results. (a) $R = 3 \text{ nm}$. (b) $R = 5 \text{ nm}$. (c) $R = 9 \text{ nm}$.

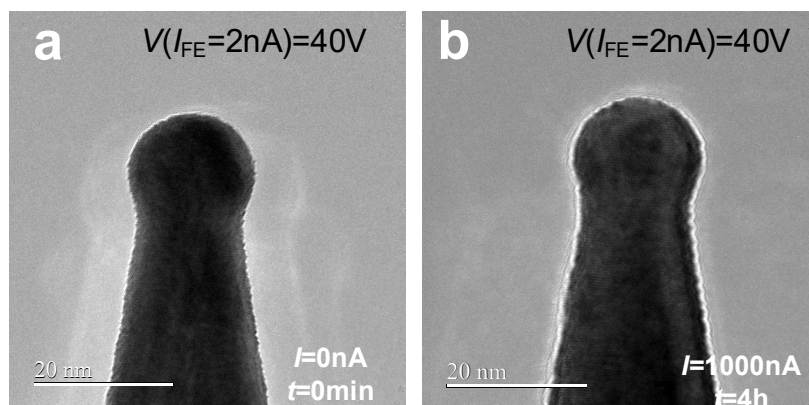


Supplementary Fig. 4| The corresponding Fourier-filtered images (to Fig. 1) of a $\langle 110 \rangle$ -oriented tungsten nanotip with an apex radius of 3 nm depict the evolution of dislocations, with the edge dislocations of different polarities marked by magenta and yellow ‘ $\perp$ ’ symbols, respectively. (a) $t = 0$ min. (b) $t = 5$ min. (c) $t = 10$ min. (d) $t = 15$ min. (e) $t = 20$ min. (f) $t = 25$ min. (g) $t = 30$ min. (h) $t = 40$ min.



Supplementary Fig. 5| Field emission I - V curves of tungsten nanotips. (a) $R = 3$ nm. (b) $R = 5$ nm. Source data are provided as a Source Data file.

38



39

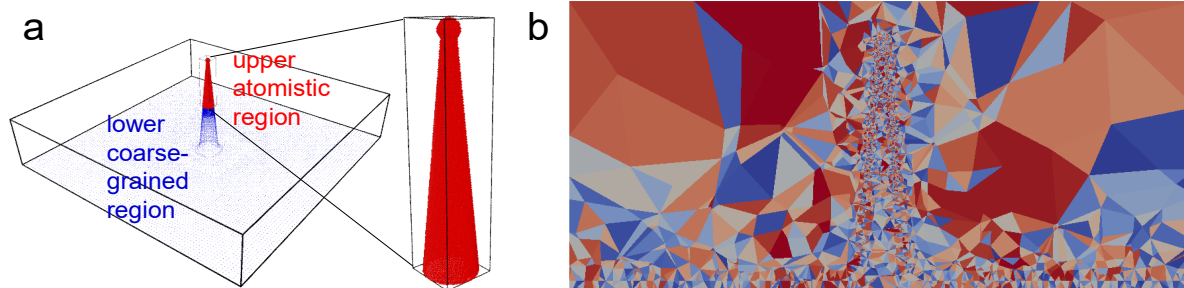
40 **Supplementary Fig. 6| The deformation behaviors of tungsten nanotip with an apex radius of 9 nm.**

41 (a) The initial TEM image of tungsten nanotip. (b) The TEM image of this tungsten nanotip after field
 42 emission treatment with $I_{\text{FE}} = 1000\text{ nA}$ and $\Delta t = 4\text{ h}$. The applied voltages required to achieve a field
 43 emission current of 2 nA ($d = 15\text{ nm}$) are provided.

44

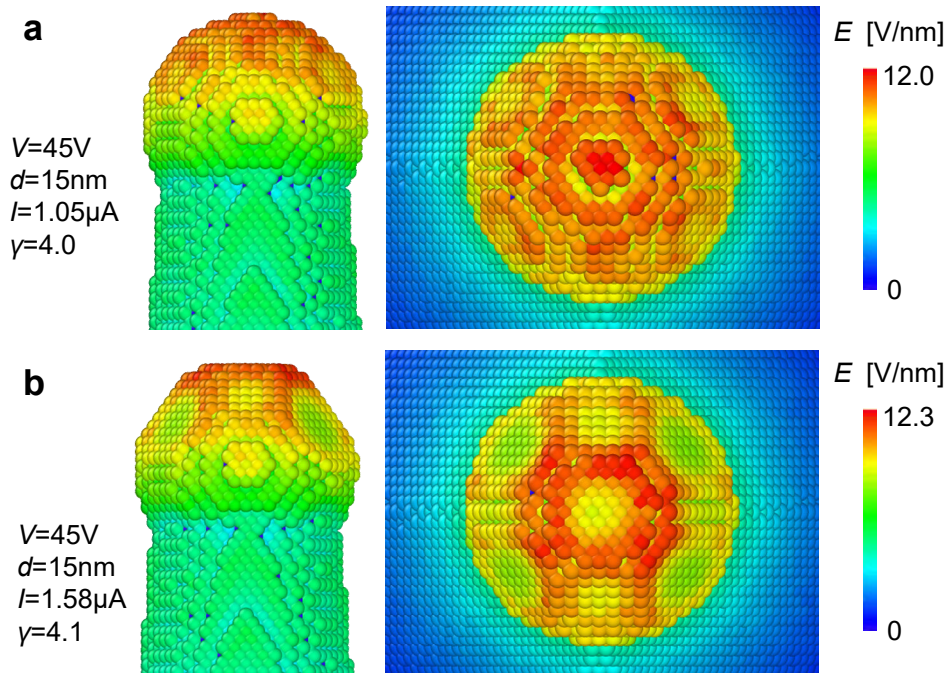
45 **Supplementary Note 1: The simulated electric field distribution for atomistic models**

46 Based on the established model (a typical example is shown in Supplementary Fig. 7), the
 47 atomically mapped field distributions of spherical and polyhedral nanotips with apex radii of 3 nm,
 48 5 nm, and 9 nm are shown in Supplementary Figs. 8-10. It can be seen that the electric field is
 49 stronger at the edge atoms, and the polyhedral nanotips exhibit only a slight increase in field
 50 strength compared with the spherical ones.

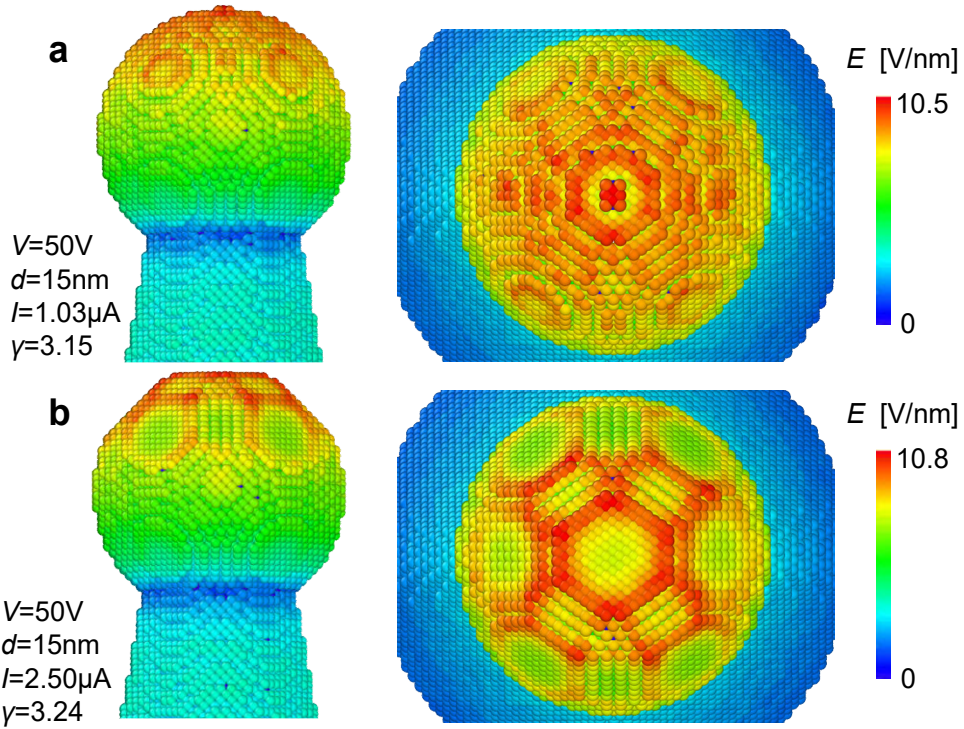


51

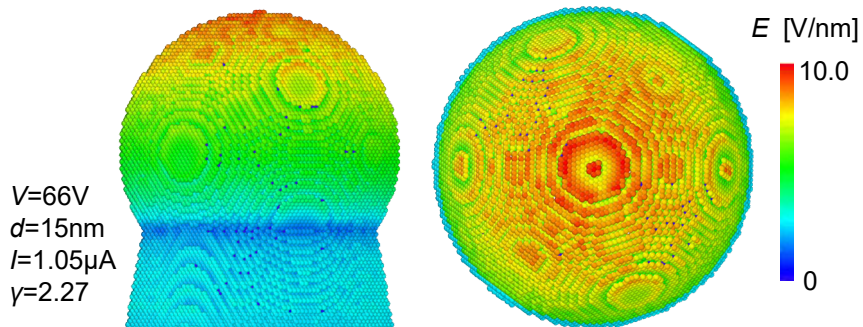
52 **Supplementary Fig. 7| The typical nanotip model in FEcMD simulation.** (a) The nanotip model. (b)
 53 The finite element mesh.



54
 55 **Supplementary Fig. 8| The electric field distribution for the atomistic spherical nanotip and the**
 56 **polyhedral nanotip with a 3 nm apex radius.** (a) The spherical nanotip. (b) The polyhedral nanotip. On
 57 the left side of the figure, the applied voltage across the gap, the resulting field emission current, and the
 58 field enhancement factor obtained from the simulation are shown.



Supplementary Fig. 9| The electric field distribution for the atomistic spherical nanotip and the polyhedral nanotip with a 5 nm apex radius. (a) The spherical nanotip. (b) The polyhedral nanotip. On the left side of the figure, the applied voltage across the gap, the resulting field emission current, and the field enhancement factor obtained from the simulation are shown.



Supplementary Fig. 10| The electric field distribution for the atomistic spherical nanotip with a 9 nm apex radius. Side and top views of the field distribution are shown. On the left side of the figure, the applied

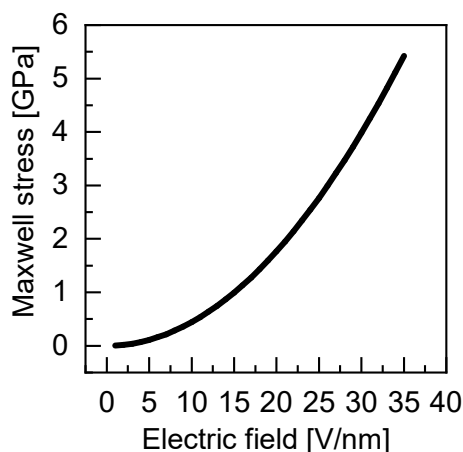
voltage across the gap, the resulting field emission current, and the field enhancement factor obtained from the simulation are shown.

Supplementary Note 2: The maxwell stress induced by electric fields

The most significant effect of an external electric field on a metal surface is that it exerts an a tensile stress to the material surface, i.e. the Maxwell stress σ

$$\sigma = \frac{\epsilon_0}{2} F^2$$

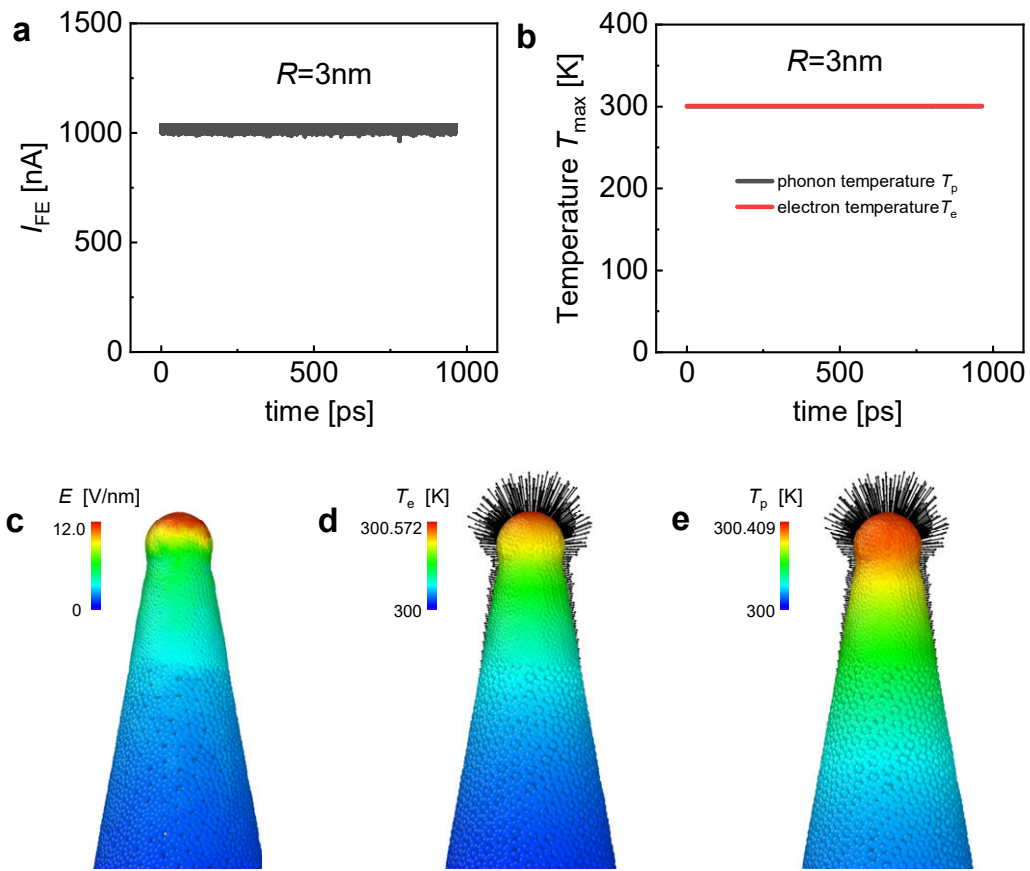
Where F is the electric field strength at a surface point and ϵ_0 is the vacuum permittivity. The Supplementary Fig. 11 shows the relationship between the Maxwell stress σ and the electric fields. The field enhacenment factor is defined as $\gamma = F_{\max}/F_{\text{avg}}$, where $F_{\text{avg}} = V/d$ is applied electric field, and $F_{\max}$ is the maximum electric field. Based on the atomistic electric field distribution obtained from FEcMD simulations (Supplementary Figs. 8-10), the field enhancement factor γ is about 4.0 for the spherical tip and 4.1 for the polyhedral tip with a 3 nm apex radius at a 15 nm gap, and γ is about 3.15 for the spherical tip and 3.24 for the polyhedral tip with a 5 nm apex radius at a 15 nm gap. Thus, the maximum electric fields $F_{\max}$ in our experiments are near 10 V/nm, corresponding to a Maxwell stress σ of less than 0.5 GPa. This value is significantly lower than the yield strength of tungsten along the $\langle 110 \rangle$ direction, which is 19.2 GPa¹. Therefore, the deformation cannot be attributed to the plastic deformation induced by the applied electric fields.



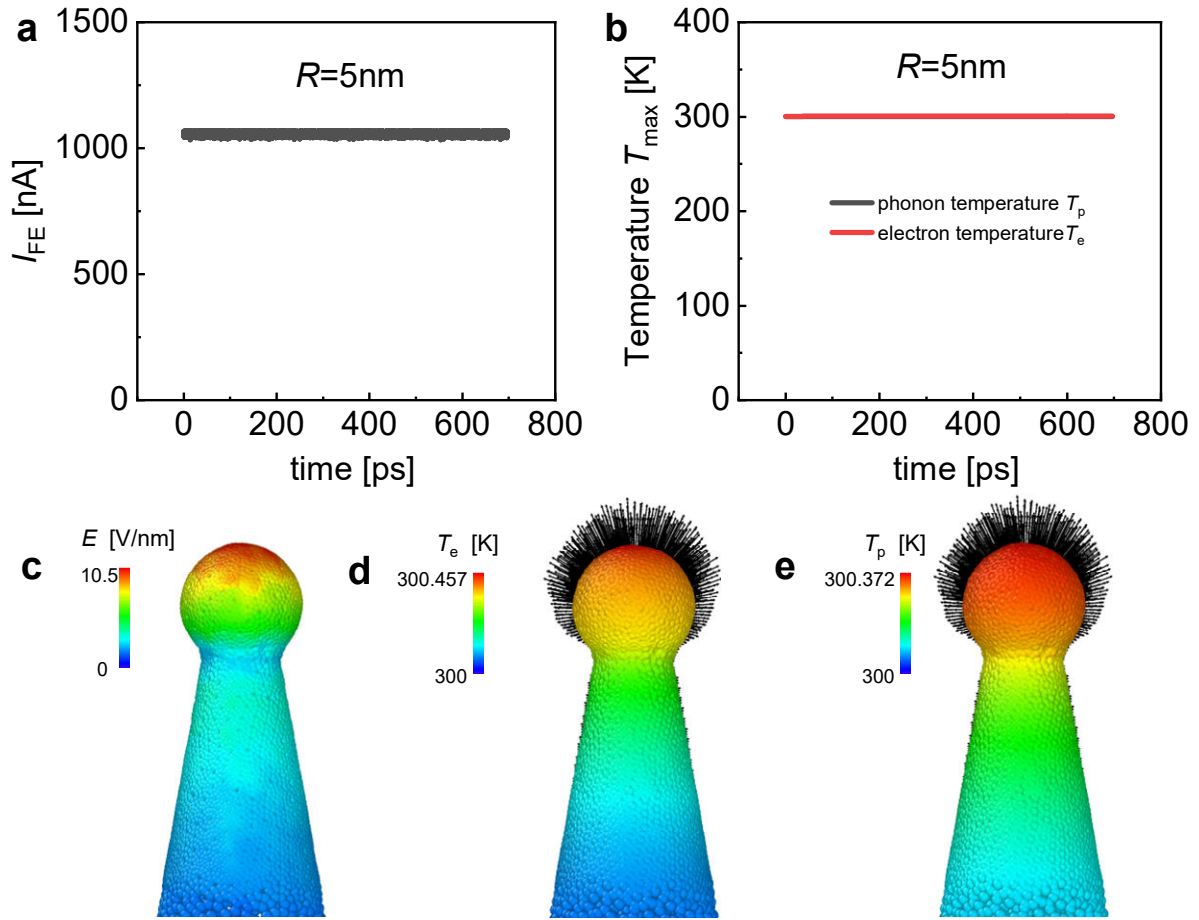
Supplementary Fig. 11| Maxwell stress induced by different electric field strengths. Source data are provided as a Source Data file.

Supplementary Note 3: The thermal effects caused by field emission

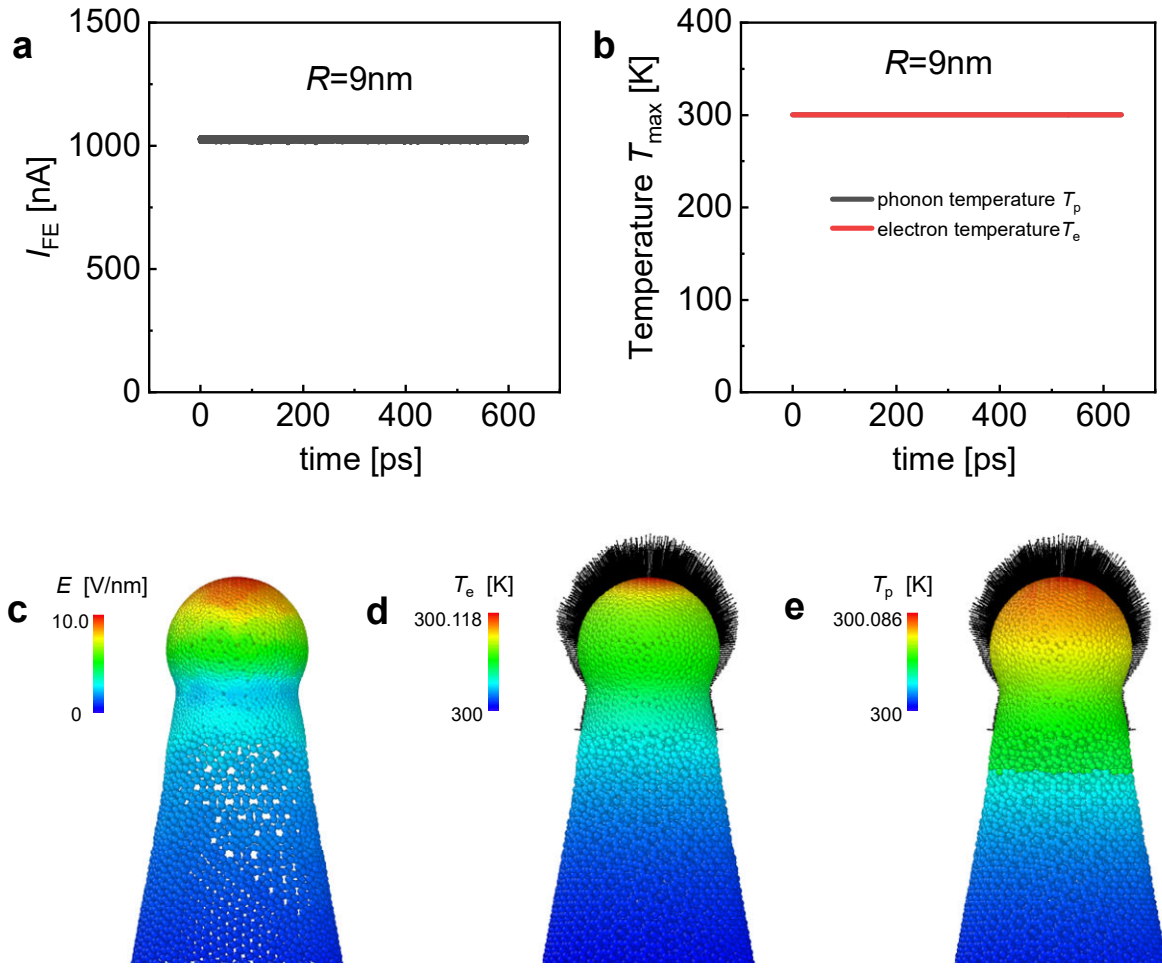
To determine the temperature of the nanotips during field emission and assess whether thermal effects significantly influence their deformation behaviors, we simulated the temperature distribution for all three nanotips under a field emission current of 1000 nA. The simulated results, including field emission current, maximum electron and phonon temperature, electric field distribution, as well as electron and phonon temperature distribution, are shown in Supplementary Figs. 12-14. When the electron and phonon temperatures stabilize and converge, this temperature can be considered the steady-state value. The simulations reveal that for all three nanotips, both electron and phonon temperatures remain around 300 K, indicating that the field emission current of about 1000 nA in the experiments does not cause a significant temperature rise.



Supplementary Fig. 12| Thermal effects of a tungsten nanotip with an apex radius of 3 nm during field emission treatment. (a) Field emission current. (b) Maximum electron temperature and phonon temperature under field emission. (c) The electric field distribution. (d) The electron temperature distribution and the electric field lines. (e) The phonon temperature distribution and the electric field lines. Source data are provided as a Source Data file.



Supplementary Fig. 13| Thermal effects of a tungsten nanotip with an apex radius of 5 nm during field emission treatment. (a) Field emission current. (b) Maximum electron temperature and phonon temperature under field emission. (c) The electric field distribution. (d) The electron temperature distribution and the electric field lines. (e) The phonon temperature distribution and the electric field lines. Source data are provided as a Source Data file.

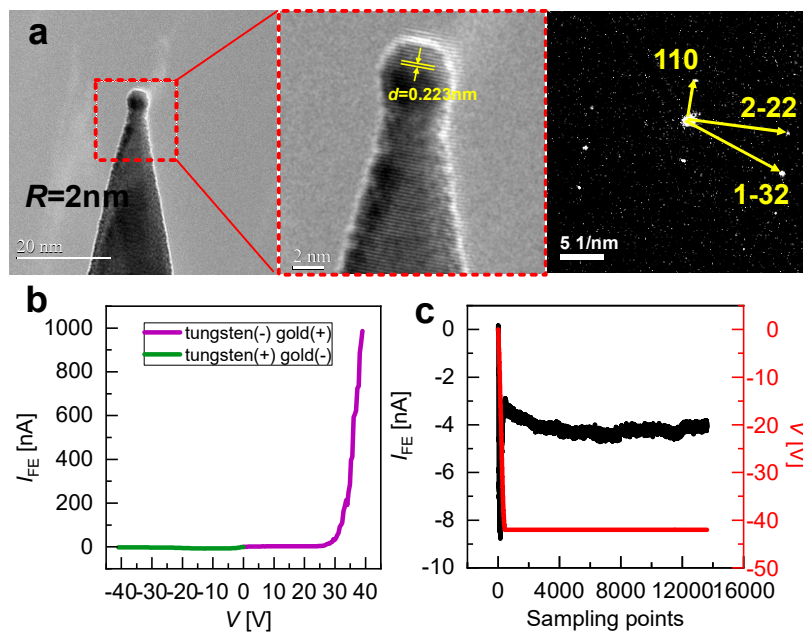


Supplementary Fig. 14| Thermal effects of a tungsten nanotip with an apex radius of 9 nm during field emission treatment. (a) Field emission current. (b) Maximum electron temperature and phonon temperature under field emission. (c) The electric field distribution. (d) The electron temperature distribution and the electric field lines. (e) The phonon temperature distribution and the electric field lines. Source data are provided as a Source Data file.

Supplementary Note 4: The electric field effects for the deformation behaviors of tungsten nanotips

To decouple the electric field effects from field emission, we conducted the reverse polarity experiments on tungsten nanotips with radii of 2 nm and 5 nm. The corresponding TEM images

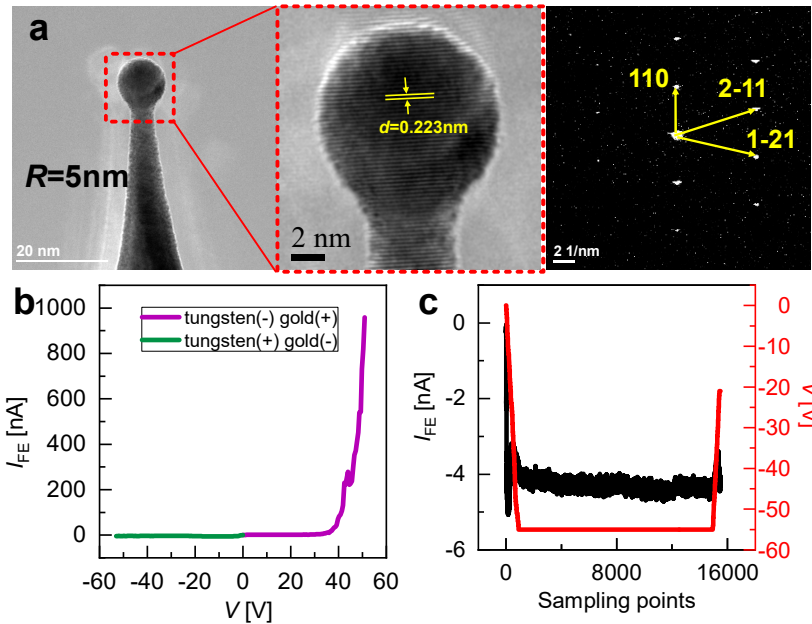
and post-processed SAED results are shown in Supplementary Fig. 15a and Supplementary Fig. 16a, which reveal a surface devoid of contaminants, a round and smooth apex, a neatly arranged lattice with few internal defects and a $\langle 110 \rangle$ crystallographic direction aligned with the emitter axis. The field emission I - V curves and the reverse polarity I - V curves are shown in Supplementary Fig. 15b and Supplementary Fig. 16b. It can be seen that the current during reverse polarity measurements is approximately 4 nA under the same electric field value, which is significantly lower than 1000 nA observed during field emission measurements, indicating successful decoupling of electric field effects from field emission. The representative voltage and current curves during reverse polarity treatment periods are shown in Supplementary Fig. 15c and Supplementary Fig. 16c.



Supplementary Fig. 15| Reverse polarity experiment of a tungsten nanotip with an apex radius of 2 nm. (a) The TEM images of this tungsten nanotip and the corresponding post-processed SAED result. (b) I - V curves for field emission characteristics (tungsten nanotip as cathode) and reverse polarity (tungsten

nanotip as anode). (c) Representative voltage and current curves during reverse polarity treatment periods.

Source data are provided as a Source Data file.



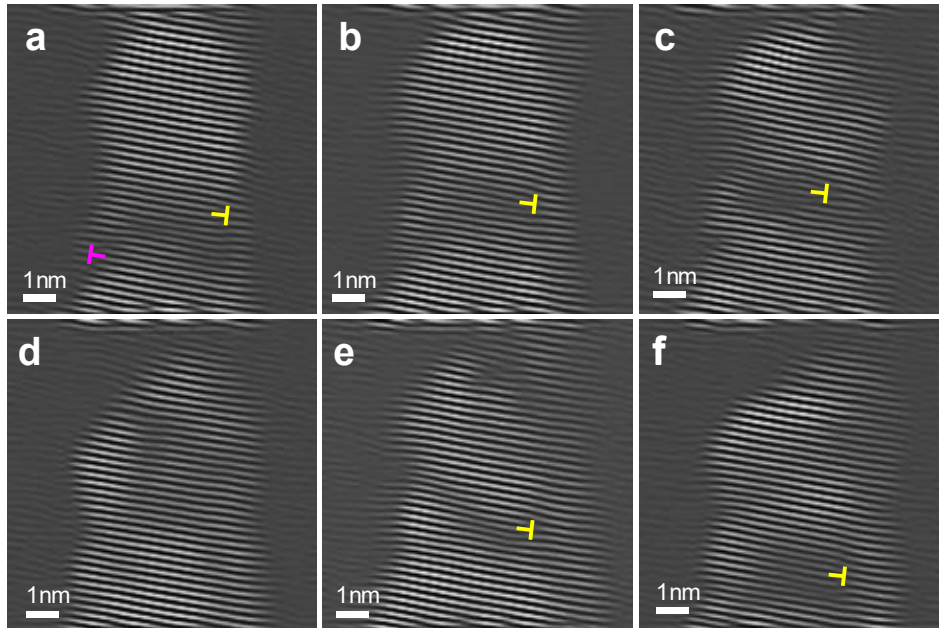
Supplementary Fig. 16| Reverse polarity experiment of a tungsten nanotip with an apex radius of 5

nm. (a) The TEM images of this tungsten nanotip and the corresponding post-processed SAED result. (b)

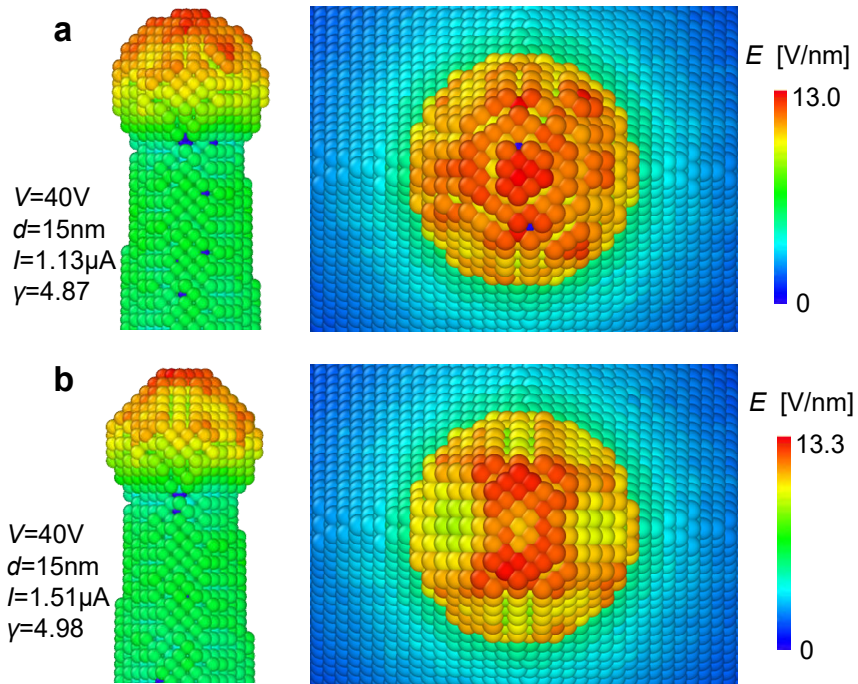
I-V curves for field emission characteristics (tungsten nanotip as cathode) and reverse polarity (tungsten

nanotip as anode). (c) Representative voltage and current curves during reverse polarity treatment periods.

Source data are provided as a Source Data file.



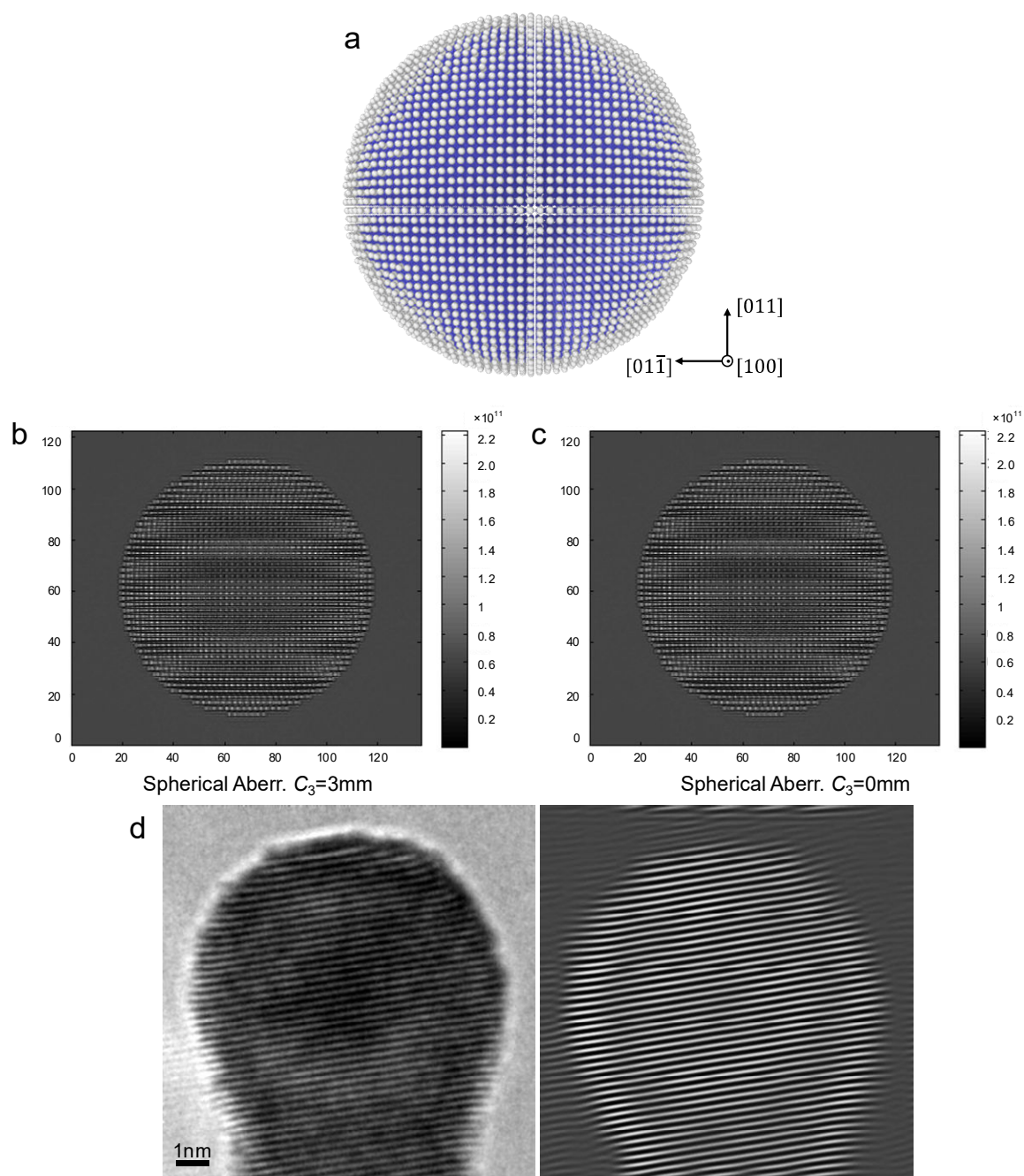
Supplementary Fig. 17| The corresponding Fourier-filtered images (to Fig. 3) of a <110>-oriented tungsten nanotip with an apex radius of 2 nm depict the evolution of dislocations, with the edge dislocations of different polarities marked by magenta and yellow 'L' symbols, respectively. (a) $t = 0$ min. (b) $t = 5$ min. (c) $t = 15$ min. (d) $t = 25$ min. (e) $t = 35$ min. (f) $t = 40$ min.



Supplementary Fig. 18| The electric field distribution for the atomistic spherical nanotip and the polyhedral nanotip with a 2 nm apex radius. (a) The spherical nanotip. (b) The polyhedral nanotip. On the left side of the figure, the applied voltage across the gap, the resulting field emission current, and the field enhancement factor obtained from the simulation are shown.

Supplementary Note 5: Multislice image simulation

The TEM images are simulated using QSTEM software with the multislice method². Two 5 nm tungsten sphere atomistic models are constructed, without or with dislocations, with the ideal zone axis along $\langle 100 \rangle$, as shown in Supplementary Fig. 19a and Supplementary Fig. 20a-b. In practice, however, the TEM holder is a single-tilt TEM holder, making it difficult to perfectly align with the zone axis, resulting in a few degrees of deviation. In our simulations, the imaging direction is set with a 7° tilt from the $\langle 100 \rangle$ zone axis. The probe array is set to 500×500 pixels, with an imaging resolution of 0.274×0.246 Å. In the slicing, the number of slices per sub-slab is 200. The microscope parameters are: high voltage 200 kV, defocus 50 nm, and spherical aberration $C_3 = 3$ mm and 0 mm. The simulated results are shown in Supplementary Fig. 19b-c and Supplementary Fig. 20c-d. As shown, for the model without dislocations, the simulated images are consistent with the FFT-filtered image in Supplementary Fig. 19d (Fig. 2a), and the spherical aberration values do not significantly affect the imaging. For the model with dislocations, the simulated images are consistent with the FFT-filtered images with dislocations in Supplementary Fig. 20e (Fig. 2b), and the spherical aberration values also do not significantly affect the imaging. Although the number of dislocations observed from the FFT-filtered images is lower than the actual number in the nanotip, this does not affect our main conclusions.



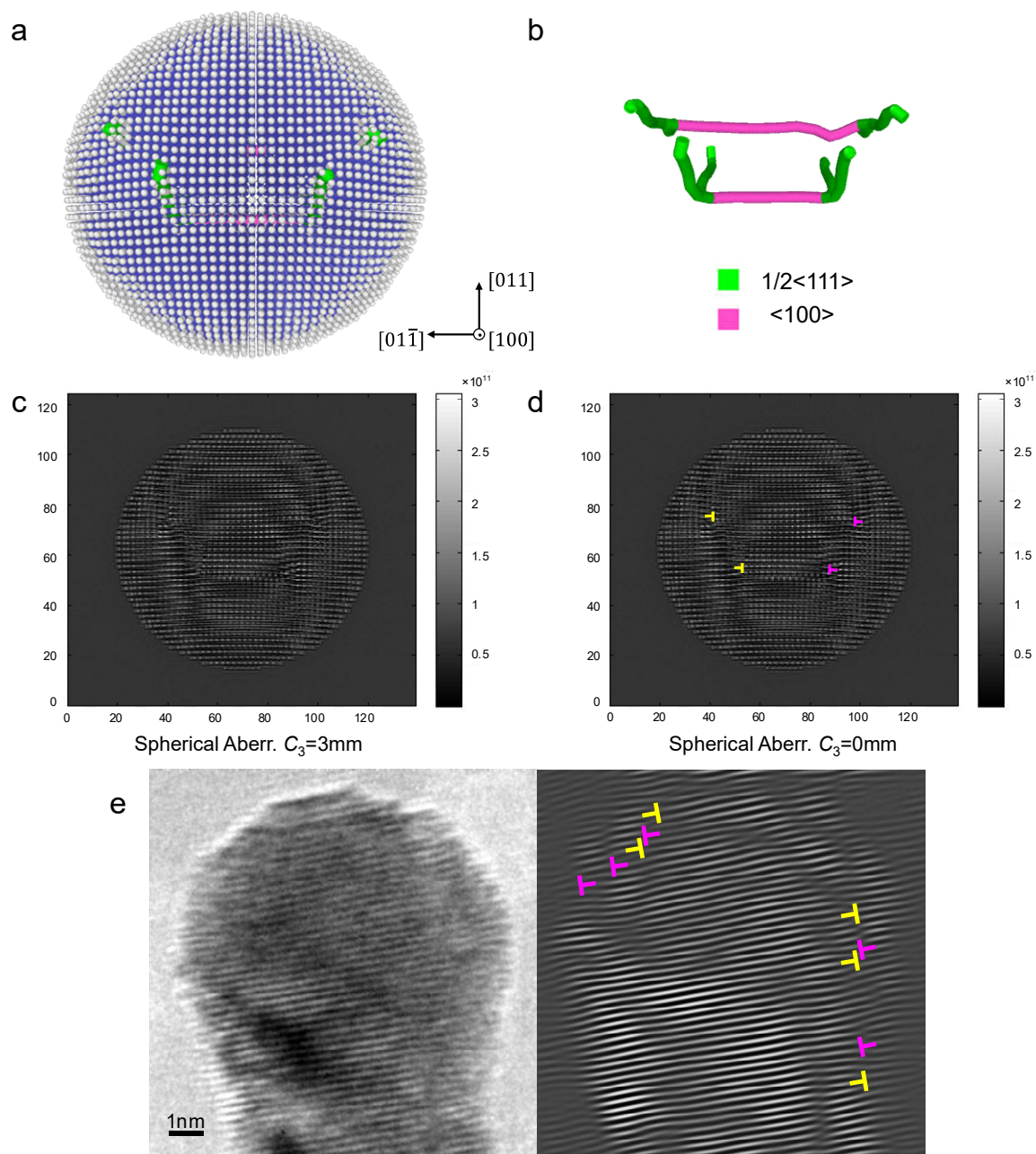
180

181 **Supplementary Fig. 19| Multislice simulation results of a 5 nm tungsten sphere without dislocations.**

182 (a) The atomistic model. (b) Simulated TEM image under spherical aberration with $C_3 = 3$ mm. (c)

183 Simulated TEM image under spherical aberration with $C_3 = 0$ mm. (d) TEM image of a 5 nm tungsten
 184 nanotip without dislocations, along with the corresponding Fourier-filtered image.

185



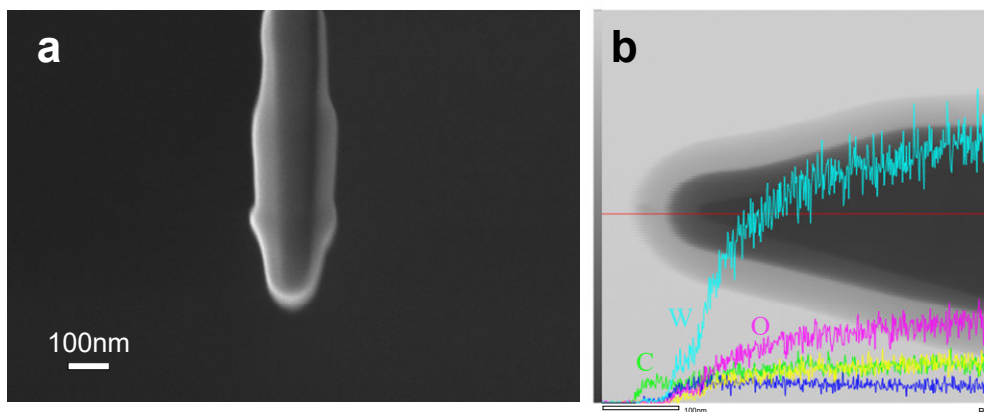
186

Supplementary Fig. 20| Multislice simulation results of a 5 nm tungsten sphere with dislocations. (a) The atomistic model. (b) The dislocations in the model. (c) Simulated TEM image under spherical aberration with $C_3 = 3$ mm. (d) Simulated TEM image under spherical aberration with $C_3 = 0$ mm. (e) TEM image of a 5 nm tungsten nanotip with dislocations, along with the corresponding Fourier-filtered image.

Supplementary Note 6: The electron beam effects of TEM on the deformation behavior of nanotip

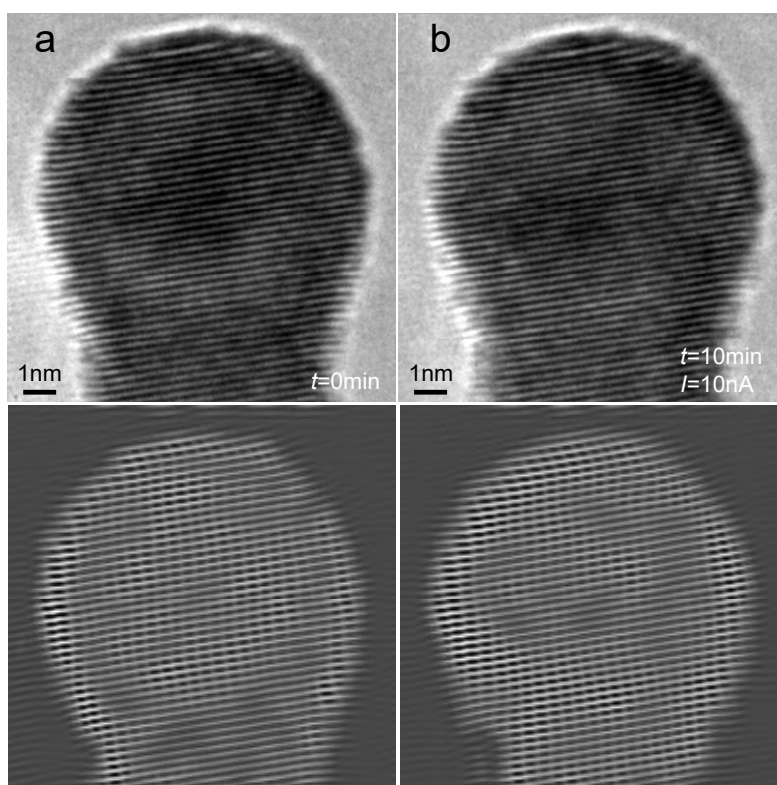
Under TEM, the electron beam may induce various effects in the electrodes, such as electron-beam sputtering, beam-induced heating, elastic displacement interactions, and beam-induced hydrocarbon contamination³, which also need to be carefully considered and evaluated. In our experiments, the TEM was operated at an accelerating voltage of 200 kV (incident electron energy of 200 keV). During real-time observation of morphological evolution, the electron beam was spread to achieve a low current density (dose rate) of ~ 4 pA/cm², measured on a 160 mm-diameter screen. (i) For electron-beam sputtering of tungsten, the threshold incident electron energy is about 500 keV, and thus it can be neglected under our experimental conditions (~ 200 keV). (ii) For beam-induced heating, the temperature rise caused by electron beam at a dose rate of ~ 4 pA/cm² is minimal (< 1 K)³⁻⁵. (iii) For elastic displacement interactions, the surface diffusion energy E_{sd} of tungsten is about 0.8 eV⁶, which is typically lower than the adsorption energy E_{ad} by a factor of three or more⁷. This E_{sd} value corresponds to a threshold incident electron energy of approximately 220 keV. In our experiments, the electron energy is 200 keV, which is close to this threshold, suggesting that beam-induced surface diffusion may occur. To further assess the contribution of beam-induced effects, we estimated the beam-induced surface-displacement rate (see Eq. 10 in Ref. 7), which is proportional to the beam current density. Given $E_{sd} = 0.8$ eV, $J = 4$ pA/cm², and the atomic number of tungsten $Z = 74$ (see Fig. 4 in Ref. 7), we obtain the displacement rate to be

211 on the order of 10^{-13} s^{-1} ⁷. In our experiments, although the nanotip surface contains atomic steps
212 where atoms may preferentially diffuse along step edges due to lower E_{sd} ($\sim 0.58 \text{ eV}$)⁶, a one-order-
213 of-magnitude difference (see Fig. 4 in Ref. 7) in the displacement rate is insufficient to produce
214 significant beam-induced knock-on displacement to contribute surface atom diffusion⁷. This low
215 rate is also insufficient to generate the point-defect density needed to nucleate dislocations or to
216 drive the migration of pre-existing ones. This is confirmed by the clear difference in the TEM
217 images obtained for the same nanotip in both polarities of the applied field. As we can see, although
218 the beam energy is close to the threshold incident electron energy for surface diffusion, the very
219 low beam current density (far below the field emission densities $\sim 10^6 \text{ A/cm}^2$) ensures that knock-
220 on displacement can be neglected. (iv) It is also known that the beam-induced hydrocarbon
221 contamination may occur during the TEM imaging³, however, no obvious amorphous carbon layer
222 was observed during several hours of morphological evolution ($\sim 10^{-5} \text{ Pa}$ vacuum level in TEM)
223 (Fig. 2 and Fig. 4), in contrast to our earlier SEM observations at high magnification
224 (Supplementary Fig. 21), where pronounced amorphous carbon deposition occurred within
225 seconds. Moreover, during field emission, the field-emission characteristics remained relatively
226 stable, without noticeable fluctuations (see Supplementary Fig. 2 and Supplementary Fig. 5),
227 unlike our previous observation of fluctuating field-emission behavior in a carbon-coated tungsten
228 nanotip⁸. Since the tungsten tip acts as an electron source during field emission, the high current
229 density can even help to clean the tip surface^{9, 10}. Therefore, we can state with the confidence that
230 the effects of the imaging electron beam are insignificant for the morphology evolution observed
231 in our experiments.



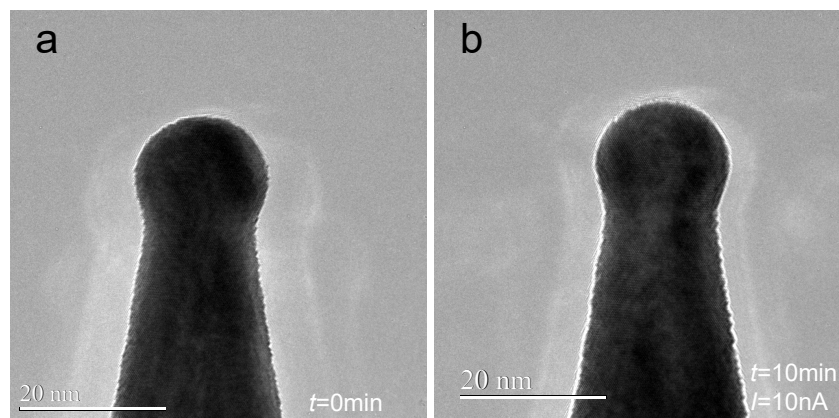
232

233 **Supplementary Fig. 21| The tungsten nanotip covered by an amorphous carbon layer.** (a) The tungsten
 234 nanotip (prepared by the double-electrolyte electrochemical etching method) in SEM covered by an
 235 amorphous carbon layer formed via beam-induced hydrocarbon contamination. (b) The corresponding EDS
 236 line scan along the axis of representative nanotip covered by an amorphous carbon layer.



237

Supplementary Fig. 22| The 5 nm tip morphology before and after 10 nA field emission current treatment. (a) $t = 0$ min (before field emission current treatment). (b) $t = 10$ min (after 10 nA field emission current treatment).



Supplementary Fig. 23| The 9 nm tip morphology before and after 10 nA field emission current treatment. (a) $t = 0$ min (before field emission current treatment). (b) $t = 10$ min (after 10 nA field emission current treatment).

Supplementary Reference

1. Wang, J. W., Zeng, Z., Weinberger, C. R., Zhang, Z., Zhu, T., Mao, S. X. atomic-scale observation of twinning-dominated deformation in nanoscale body-centred cubic tungsten. *Nat Mater* **14**, 594-600 (2015).
2. Koch, C. T. Determination of core structure periodicity and point defect density along dislocations. Arizona State University (2002).
3. Egerton, R. F., Li, P., Malac, M. Radiation damage in the TEM and SEM. *Micron* **35**, 399-409 (2004).
4. Sun, J., He, L. B., Lo, Y. C., Xu, T., Bi, H. C., Sun, L. T., Zhang, Z., Mao, S. X., Li, J. Liquid-like pseudoelasticity of sub-10-nm crystalline silver particles. *Nat Mater* **13**, 1007-1012 (2014).
5. Williams, D. B., Carter, C. B. The transmission electron microscope (Springer, New York, 1996).
6. Yang, J. Y., Hu, W. Y., Tang, J. F. Surface self-diffusion behavior of individual tungsten adatoms on rhombohedral clusters. *J Phys-Condens Mat* **23**, 395004 (2011).
7. Egerton, R. F. Beam-Induced Motion of Adatoms in the Transmission Electron Microscope. *Microsc Microanal* **19**, 479-486 (2013).

- 263 8. Meng, G., Li, Y., Koitermaa, R. A., Zadin, V., Cheng, Y., Kyritsakis, A. In Situ
264 Observation of Field-Induced Nanoprotrusion Growth on a Carbon-Coated Tungsten
265 Nanotip. *Phys Rev Lett* **132**, 176201 (2024).
- 266 9. de Parga, A. L. V., Hernan, O. S., Miranda, R., Yeyati, A. L., Mingo, N., Martin-Rodero,
267 A., Flores, F. Electron resonances in sharp tips and their role in tunneling spectroscopy.
268 *Phys Rev Lett* **80**, 357-360 (1998).
- 269 10. Feenstra, R. M., Stroscio, J. A., Fein, A. P. Tunneling Spectroscopy of the Si(111)2x1
270 Surface. *Surf Sci* **181**, 295-306 (1987).

271