Supplemental Information

## 1. Principle of electron holography

According to the basic theory of electron holography, the hologram is a series of interference fringes caused by the electron waves divided by the biprism in TEM, and the reconstructed phase map is actually the phase difference of the two waves. In this research, the phase distribution of the reconstructed image including LLZO and Pt electrode without bias $\varphi^0(x,y)$ is determined by the phase difference between the objective phase $\varphi_{obj}(x,y)$ and the reference phase involving $\varphi_{\mathrm{vac}}(x,y)$. $\varphi_{obj}(x,y)$ contains the mean inner potential and the possible dynamic diffraction effect while $\varphi_{\mathrm{vac}}(x,y)$ is the intrinsic phase of electron wave pass through the vacuum, which is assumed as constant

$$\varphi^0(x,y) = \varphi_{obj}(x,y) - \varphi_{\mathrm{vac}}(x,y) \tag{1}$$

When a bias is applied to the gate, the phase $\varphi^{bias}(x,y)$ should be changed to

$$\varphi^{bias}(x,y) = \varphi_{obj}^{\mathrm{bias}}(x,y) + \varphi_{obj}(x,y) - \varphi_{\mathrm{vac}}(x,y) \tag{2}$$

where $\varphi_{obj}^{\mathrm{bias}}(x,y)$ is the phase shift caused by the accumulated charges. Using Eq. (1) and (2), there is

$$\varphi_{obj}^{\mathrm{bias}}(x,y) = \varphi^{bias}(x,y) - \varphi^0(x,y) = \mathrm{C}_E V^{bias}(x,y)t \tag{3}$$

where $C_E$ = 0.0073 V$^{-1}$ nm$^{-1}$ for electron wave accelerated at 200 kV, $t$ is the sample thickness and $V^{bias}(x,y)$ is the potential relating the charges induced by external bias. Thus, the retrieved "net" phase maps $\varphi_{obj}^{\mathrm{bias}}(x,y)$ can reflect the charge distribution directly.

## 2. SAED in the in situ experiments

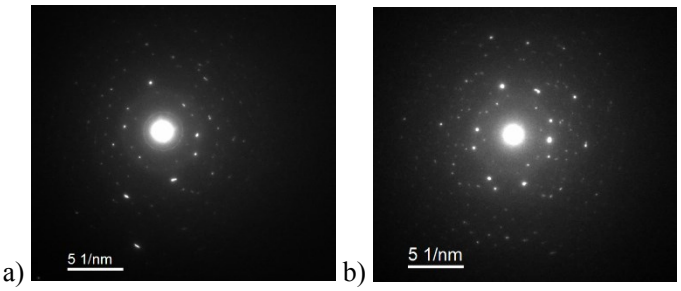



Fig. s1 a) and b) The original SAED of the pristine and charged LLZO in Fig. 6b, respectively.

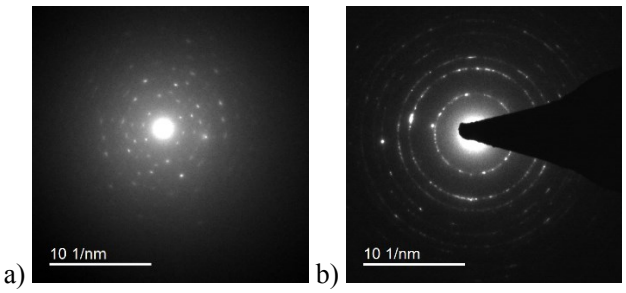



Fig. s2 a) and b) The original SAED of the charged LLZO and Li extrusion in Fig. 1c, respectively.

2. 2. Extraction of Li during the in situ operation

SI_v1 1 is the original speed record in which the screen shots are shown in Fig. 6a. Fig. s3 shows the screen shots from SI_v2 of another operation, which is accelerated four times.

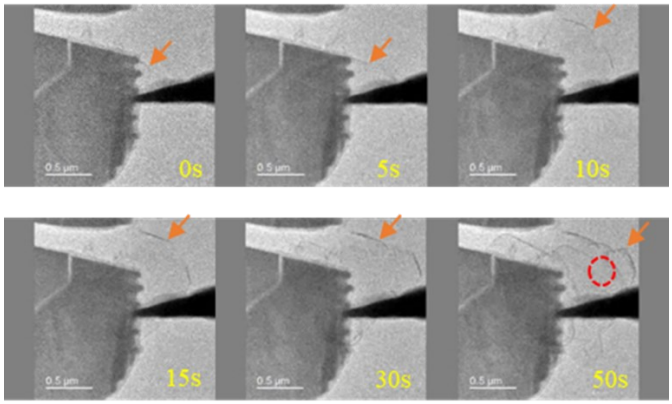



Fig. s3 The screen shots from SI_v2 shows the Li extrusion on -40 V bias. Scale bar is 500 nm.

3. EELS

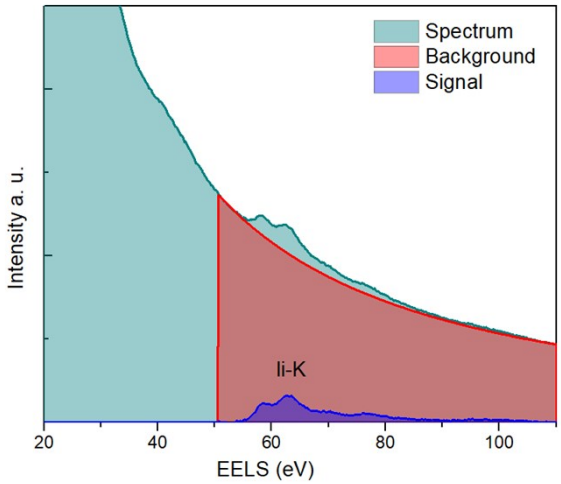
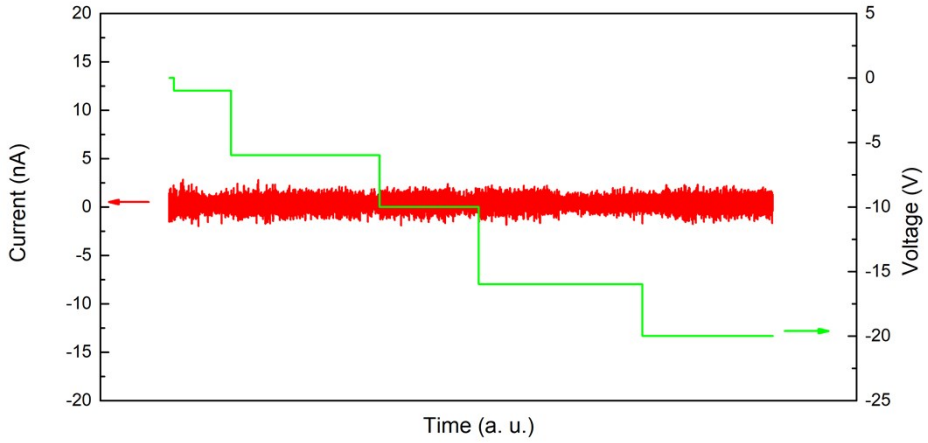
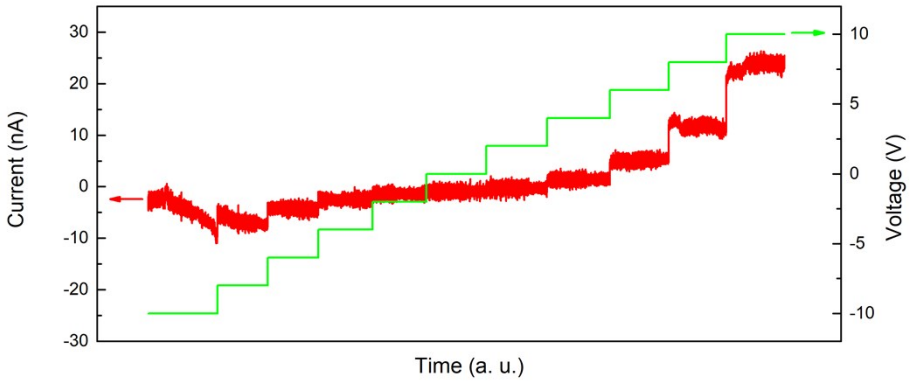



Fig. s3 The EELS of the extrusion under -40 V bias shows the evident Li signal.

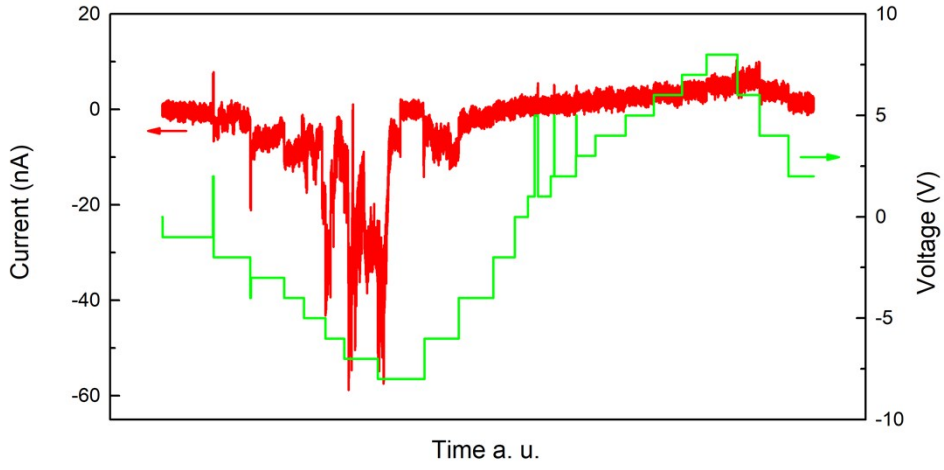
4. I-V curves during the in situ test




a)




b)




c)

Fig. S4 Current (red) and voltage (green) changes at the in-situ experiments for Figure 2, Figure 3ab, and Figure 3c in the paper, respectively.