Supporting Information

**Solid-State Electrochemical Switch of Superconductor-Metal-Insulators**

Xi Zhanga\*, Gowoon Kimb, Qian Yangb, Jiake Weic,d, Bin Fengc, Yuichi Ikuharac,d and Hiromichi Ohtaa\*

a *Research Institute for Electronic Science, Hokkaido University, N20W10, Kita, Sapporo 001-0020, Japan*

b *Graduate School of Information Science and Technology, Hokkaido University, N14W9, Kita, Sapporo 060-0814, Japan*

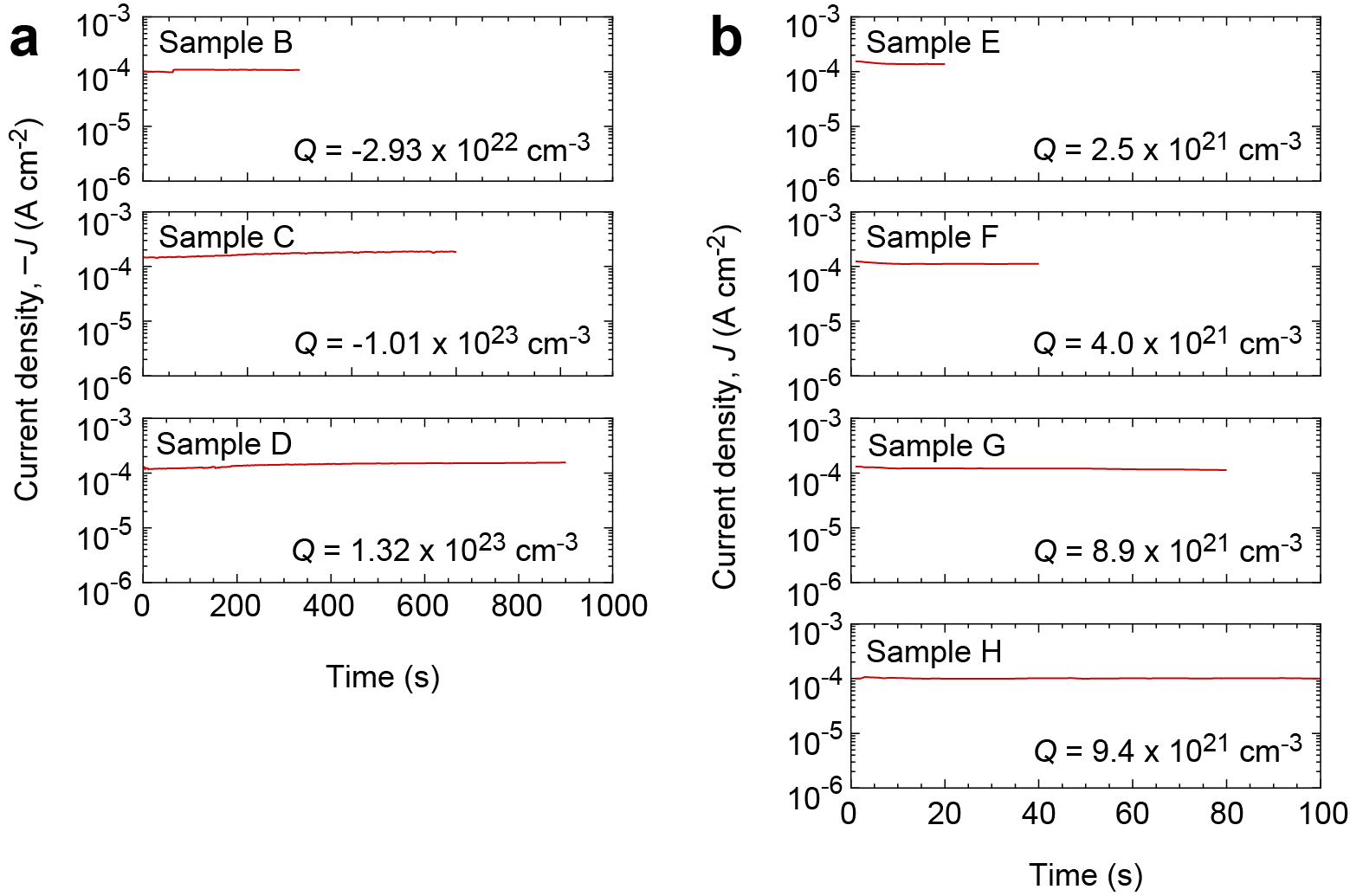
c *Institute of Engineering Innovation, The University of Tokyo, 2-11-16 Yayoi, Bunkyo, Tokyo 113-8656, Japan*

d *Elements Strategy Initiative for Structural Materials, Kyoto University, Yoshida-honmachi, Sakyo-ku, Kyoto 606-8501, Japan*

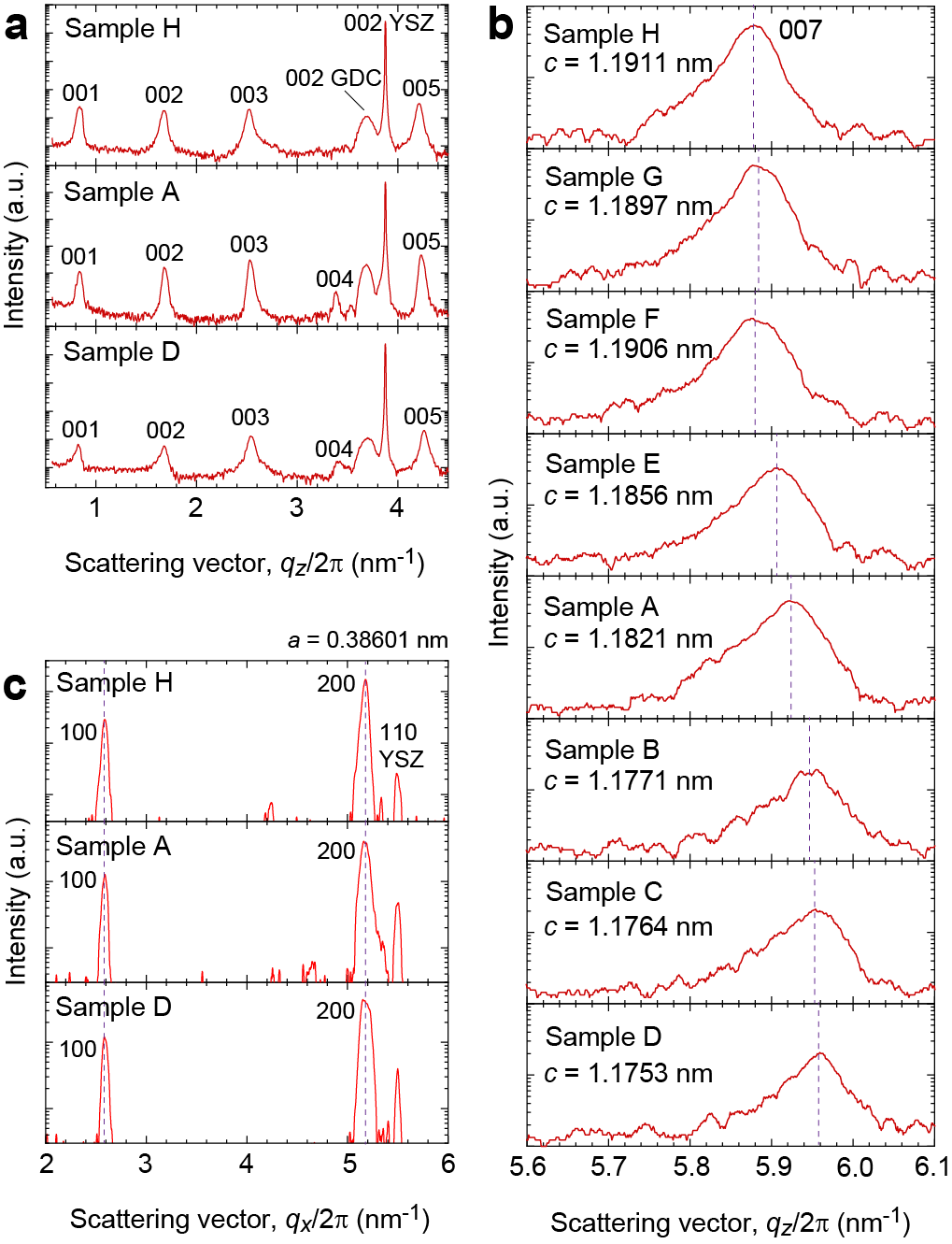
\*Email: zhangxielle@gmail.com, hiromichi.ohta@es.hokudai.ac.jp

**Table S1.** Oxygen deficiency (*δ*), electron density (*Q*), lattice parameter (*c*), thermopower (*S*) at room temperature, electrical resistivity (*ρ*) at room temperature, and superconducting transition temperature (*T*c) of the YBCO samples.

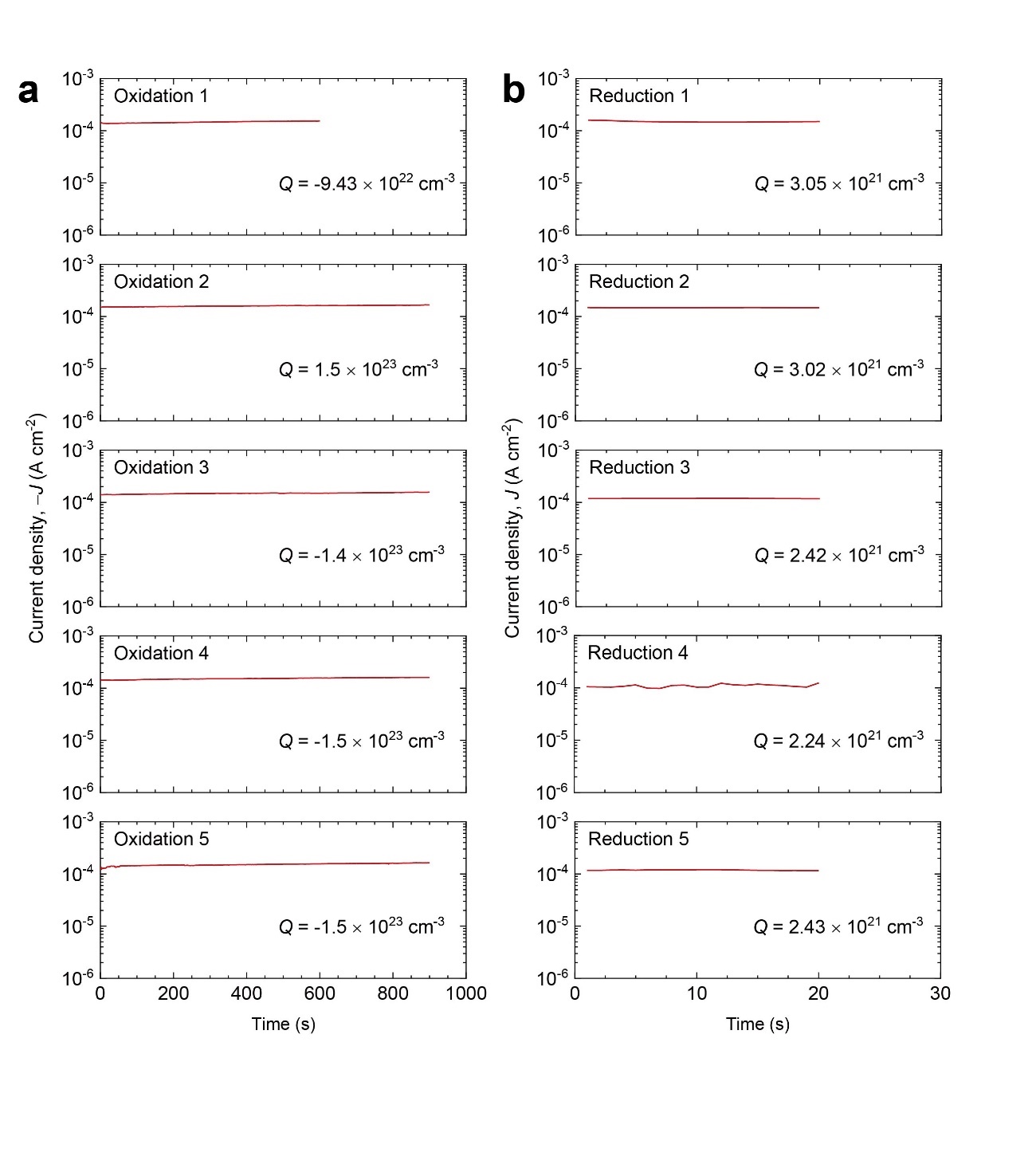
|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Sample | *δ* | *Q* (cm−3) | *c* (nm) | *S* (μV K−1) | *ρ* (Ω cm) | *T*c (K) |
| A | 0.28 | 0 | 1.1821 | 20.1 | 8.32 × 10−4 | 41 |
| B | 0.098 | −2.93 × 1022 | 1.1771 | 0.96 | 4.56 × 10−4 | 50 |
| C | 0.071 | −1.01 × 1023 | 1.1764 | −1.39 | 3.58 × 10−4 | 66 |
| D | 0.069 | −1.32 × 1023 | 1.1753 | −1.61 | 1.63 × 10−4 | 87 |
| E | 0.64 | +2.5 × 1021 | 1.1856 | 81.5 | 4.29 × 10−3 |  |
| F | 0.72 | +4.0 × 1021 | 1.1906 | 110 | 8.42 × 10−3 |  |
| G | 0.86 | +8.9 × 1021 | 1.1897 | 218 | 5.99 × 10−2 |  |
| H | 0.87 | +9.4 × 1021 | 1.1911 | 241 | 7.69 × 10−2 |  |



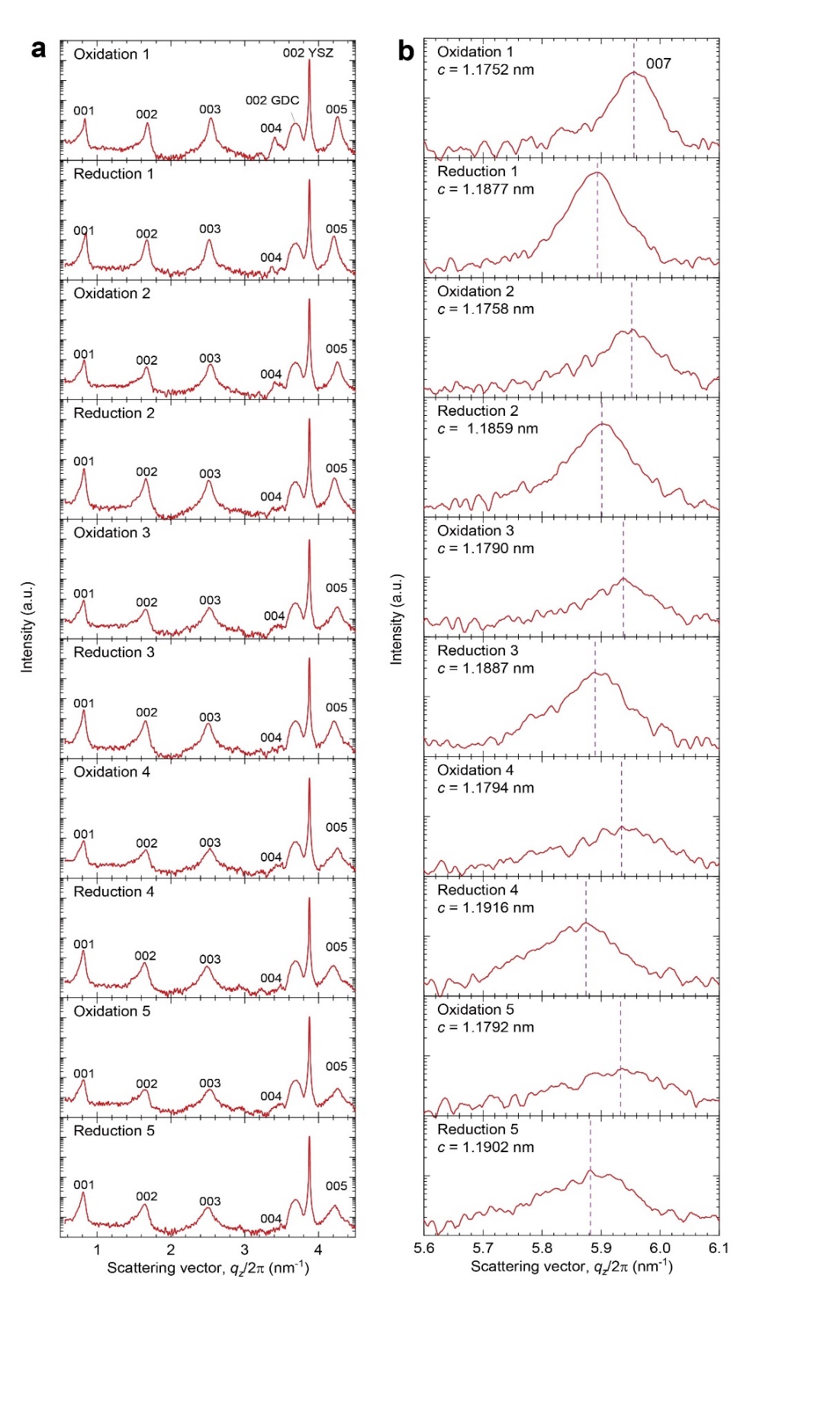
**Figure S1**. Applied current densities for the electrochemical redox treatment. (a) Oxidation treatment. (b) Reduction treatment.



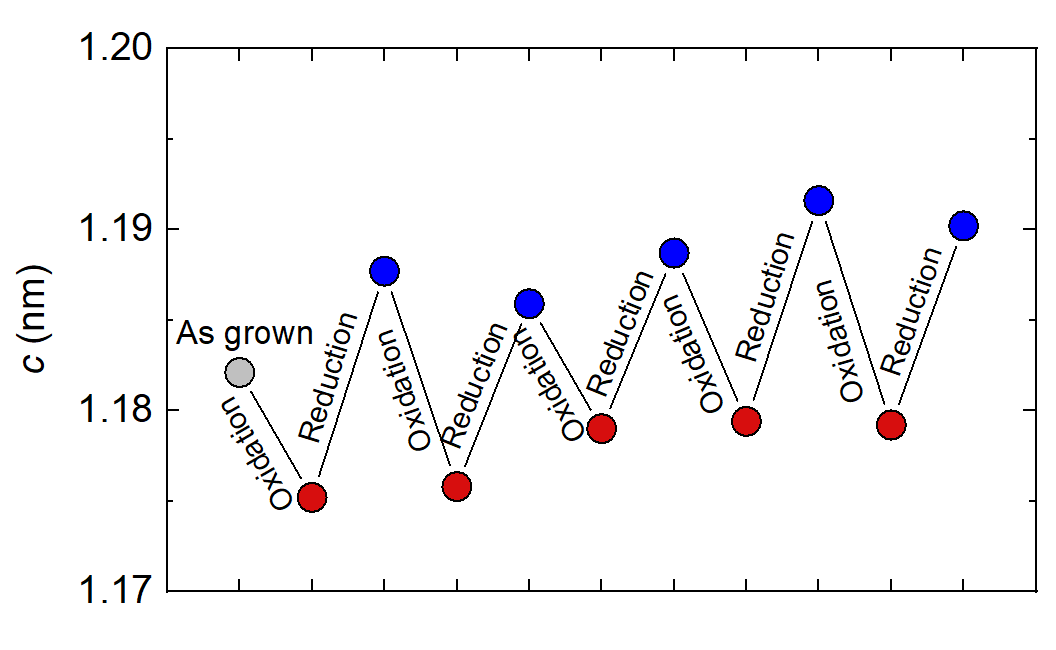
**Figure S2**. X-ray diffraction patterns of the YBa2Cu3O7−*δ* films with various redox states. (a) Out-of-plane XRD patterns. Intense diffraction peaks of 00*l* YBa2Cu3O7−*δ* are observed along with 00*l* GDC/YSZ. (b) Magnified out-of-plane XRD patterns around 007 YBCO. (c) In-plane X-ray Bragg diffraction patterns. Diffraction peak positions of samples A, D, and H are the same. *a*-axis lattice parameter of the samples is 0.38601 nm. [A: as-grown, B: *Q* = −2.93 × 1022 cm−3, C: *Q* = −1.01 × 1023 cm−3, D: *Q* = −1.32 × 1023 cm−3, E: *Q* = +2.5 × 1021 cm−3, F: *Q* = +4.0 × 1021 cm−3, G: *Q* = +8.9 × 1021 cm−3, and H: *Q* = +9.4 × 1021 cm−3]



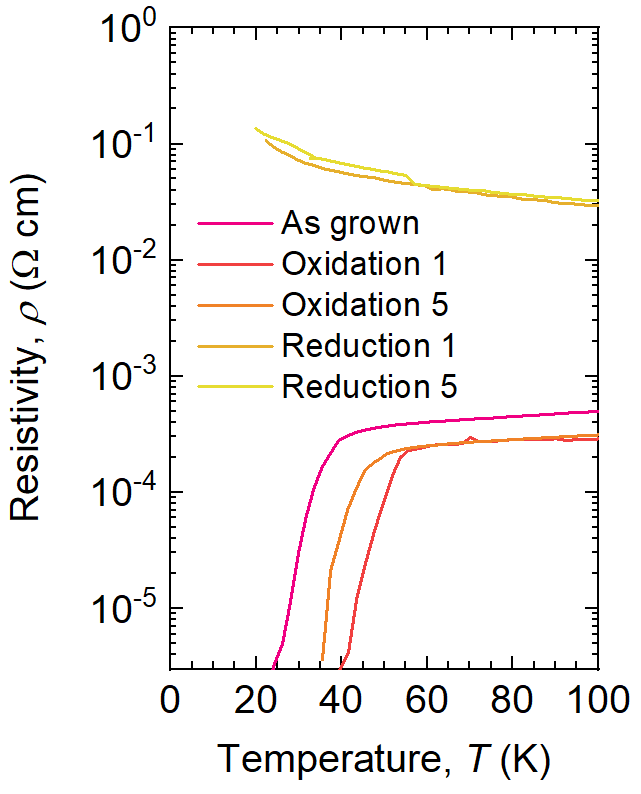
**Figure S3**. Applied current densities for the electrochemical redox treatment. (a) Oxidation treatment. (b) Reduction treatment. The oxidation and reduction treatment were alternatively applied.



**Figure S4**. X-ray diffraction patterns of the YBa2Cu3O7−*δ* films after cycling electrochemical oxidation and reduction. (a) Out-of-plane XRD patterns. Intense diffraction peaks of 00*l* YBa2Cu3O7−*δ* are observed along with 00*l* GDC/YSZ after 5 oxidation-reduction cycles. (b) Magnified out-of-plane XRD patterns around 007 YBCO. The peak positions show clear revisable shift by cycling electrochemical oxidation and reduction.



**Figure S5**. *c*-axis lattice parameter changes after cycling electrochemical oxidation and reduction.



**Figure S6**. *ρ*−*T* curves of the YBa2Cu3O7−*δ* films after different oxidation-reduction cycles. The superconductor-metal-insulator modulation shows good cyclability by repeating the electrochemical redox treatment for 5 times.