Supporting Information for

Coulomb Explosion Strategy to Tailor Nano-architecture of α-MoO₃ Nanobelts and Insight of its Intrinsic Mechanism

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Figure S1. Bandgap analysis of the α -MoO₃ nanobelts. UV-visible diffuse absorbance spectra of the α -MoO₃ nanobelts. The bandgap of the α -MoO₃ nanobelts can be estimated by the UV-visible adsorption results,^[1] which shows that the bandgap value is about 3.1 eV. It demonstrates the semiconductor properties of α -MoO₃ nanobelts with high resistivity, and it is possible to be engineered by the Coulomb explosion.



Figure S2. Nanopattern tailored on the α -MoO₃ NB by EBI. (a) Representative TEM image of an ultrathin α -MoO₃ NB; (b) TEM image of three gaps tailored on the NB with 100 nm lateral size.



Figure S3. Thickness and chemical analysis of the peeled-off α -MoO₃. (a) EELS low loss spectra of the peeled-off layers, which thickness can be calculated from the ratio of zero-loss peak and plasmonic peak; (b) EELS core loss spectra of the peeled-off layeres recorded with a energy dispersion of 0.25 eV/pixel to highlight the peak position and shape of the Mo-M_{4,5}, Mo-M_{2,3} and O-K edges; The energy-loss near-edge structures (ELNES) of the Mo-M_{2,3} edges (c) and O-K edges (d).

Thickness and chemical analysis of the peeled-off α -MoO₃

Thickness calculation. The thickness of TEM specimen (*e.g.*, the thickness of the peeled-off layer, *t*) can be calculated by a straightforward integration of the EELS low loss spectrum. The local thickness *t* calculation based on the log-ratio formula is expressed as:^[2]

$$t = \lambda \ln(I_t / I_0) \tag{1}$$

where, I_t and I_0 are the total and zero-loss areas in the low-loss region of EELS spectrum, respectively. λ is the total inelastic mean free path, which can be formulated as:

$$\lambda = 106F(E_0 / E_m) / \ln(2\beta E_0 / E_m)$$
⁽²⁾

It is noted that $F = (1 + E_0 / 1022) / (1 + E_0 / 511)^2$, E_0 is the incident energy of electron beam, β is

the spectrum collection semi-angle, $E_m = 7.6Z^{0.36}$ and Z is the atomic number. According to the above equations and the collected parameters from the EELS spectrum in **Figure S3a**, the thickness of the peeled-off α -MoO₃ structure can be calculated to be about 2.1 nm, well matching a mono-layer thickness of α -MoO₃.

Table S1. Comparison of Mo-M₃ edge, the peak difference (*i.e.*, M₃---M₂) between the Mo-M₃ edge and the Mo-M₂ edge, and the peak difference (Mo-M₃---O-K) between the Mo-M₃ edge and the O-K edge with the references of stoichiometric MoO₃ and MoO₂.

Chemical compositions	Mo-M ₃ (eV)	M_{3} $M_{2}(eV)$	Mo-M ₃ O-K (eV)
Peeled-off MoO _x	397.5	16.6	131.8
MoO ₃ ^[4]	397.8	16.8	131.6
MoO ₂ ^[4]	397.7	16.5	132.2

Chemical composition analysis. The chemical compositions of the molybdenum oxides can be determined from the analysis of O-K and Mo-M edges of EELS spectra.^[3] Figure S3b shows the EELS core-loss spectrum collected from the peeled-off materials, in which the peak positions and shapes of the Mo-M_{4,5}, Mo-M_{2,3} and O-K edges demonstrating the chemical composition of the molybdenum oxide. Herein, we select the Mo-M_{2,3} edge to quantify the element and valence because it is shaper than the Mo-M_{4,5} edge and closer to the O-K edge. The ELNES spectrum of the Mo-M_{2,3} edge in Figure S3c shows two peaks at 397.5 eV and 414.1 eV, corresponding to the M₃, M₂ edges of Mo element, respectively. The energy difference between the Mo-M₃ edge and Mo-M₂ edge is the criteria to judge the oxidation state of Mo element, which is calculated to be 16.6 eV. Besides, the peak difference between the Mo-M₃ edge and the O-K edge (peak at 529.3 eV in Figure S3d) is another criteria, which is 131.8 eV. It is clear to find that the peeled-off layer is a sub-stoichiometric molybdenum oxide (*i.e.* MoO_x , 2<x<3) with low oxygen vacancy concentration based on the references of stoichiometric MoO₃ and MoO₂ in Table S1^[4]. Thus, this result proves that the oxygen can be knocked from the α -MoO₃ NB by electron beam but with a low vacancy concentration.

REFERENCE

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