

Supporting Information for

**High-mobility flexible oxyselenide thin-film transistors
prepared by solution-assisted method**

Congcong Zhang[†], Jinxiong Wu^{†,‡,*}, Yuanwei Sun^{§,#}, Congwei Tan[⊥], Tianran Li[†],
Teng Tu[†], Yichi Zhang[†], Yan Liang[†], Xuehan Zhou[†], Peng Gao^{§,#}, Hailin Peng^{†,*}

[†]Center for Nanochemistry, Beijing Science and Engineering Center for Nanocarbons, Beijing National Laboratory for Molecular Sciences, College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, China

[‡]Tianjin Key Lab for Rare Earth Materials and Applications, Center for Rare Earth and Inorganic Functional Materials, School of Materials Science and Engineering, National Institute for Advanced Materials, Nankai University, Tianjin 300350, China

[§]International Center for Quantum Materials, Electron Microscopy Laboratory, School of Physics, Peking University, Beijing 100871, China

[#]Collaborative Innovation Center of Quantum Matter, Peking University, Beijing 100871, China

[⊥]Academy for Advanced Interdisciplinary Studies, Peking University, Beijing 100871, China

*To whom correspondence should be addressed.

Email: jxwu@nankai.edu.cn, hlpeng@pku.edu.cn.

Part-I: Methods

1. Preparation and characterizations of Bi₂O₃ thin film and Bi₂O₂Se thin film

Firstly, 1 g Bi(NO₃)₃·5H₂O (purchased from Energy Chemical, China, 99%) was dissolved in 10 mL (CH₂OH)₂ (purchased from Sinopharm Chemical Reagent, HPLC) to form colorless and transparent solution. Secondly, the Bi(NO₃)₃/(CH₂OH)₂ solution was dropped onto the muscovite with fresh cleavage plane and the solution was spin-coated at a certain rotate speed (2000 rpm, 3000 rpm etc.) for 60 s. Then the Bi(NO₃)₃/(CH₂OH)₂ thin film was heated at 180 °C for a few seconds to evaporate the solvent. Finally, the Bi(NO₃)₃ thin film was placed inside a quartz tube at the center of a horizontal tubular furnace in air to obtain Bi₂O₃ thin film. The furnace was programmed to reach 480 °C with elevation rate of ~8 °C/min and kept for 30 min.

As obtained the uniform Bi₂O₃ thin film was obtained, it can be transformed into Bi₂O₂Se thin film by in-situ selenization. The selenization took place in a home-made low-pressure chemical vapor deposition system (CVD) equipped with a 30-mm diameter quartz tube inside a horizontal tubular furnace (Lindberg/Blue M). The selenium pills (purchased from Alfa Aesar, 5N) and Bi₂O₃ thin film were separately located at the upstream and downstream heating zones. The selenium pills were heated to 230 °C and evaporated into gas. Then the Se₂ vapor was carried to the downstream by Argon and reacted with Bi₂O₃ thin film at 500 °C to obtain Bi₂O₂Se thin film. The system pressure was kept at 400 torr.

The morphology of Bi₂O₃ and Bi₂O₂Se thin film were performed by optical microscopy (OM, Olympus DX51 microscope) and atomic force microscopy (AFM, Bruker Dimension Icon with Nanoscope V controller). The Raman spectroscopy was performed on a confocal Raman spectrograph (JY Horiba HR800) under backscattering configuration, and the laser (633 nm) is focused on the sample by a 100× objective lens (NA=0.9). The crystal structures of Bi₂O₃ and Bi₂O₂Se were verified by X-ray diffraction (XRD, Rigaku D/Max-2000 diffractometer, Cu K α radiation (λ =0.15406 nm) at 40 kV and 100 mA). The cross-sectional STEM images were recorded using an aberration-corrected Titan Themis G2 microscope at 300 kV with a beam current of 20 pA. The convergence semi-angle for imaging is 30 mrad, the collection semi-angles snap is 39 to 200 mrad.

2. Device fabrication on muscovite and transfer onto flexible substrate

The Hall-bars were fabricated directly on the insulating muscovite substrate. We first patterned the Bi₂O₂Se continuous thin film (8 nm) into discrete 40×20 μm^2 sheets by standard photolithography techniques and wet chemical etching method reported before. Second, alignment marker arrays were predefined onto the muscovite with standard photolithography techniques. A standard electron beam lithography (EBL) process was then applied to create a polymethyl methacrylate (PMMA) mask that exposes source/drain region of Hall-bars for depositing Pd/Au electrodes (5/50 nm) by thermal evaporation. Top-gate field-

effect transistors (FETs) were fabricated with the same process first. The top gate dielectrics were formed through depositing 20 nm HfO₂ by atomic layer deposition (ALD), followed by depositing 5/50 nm Cr/Au as top-gate electrodes by thermal evaporation.

Part II: Results and discussion

1. Selection of substrate and precursor for Bi₂O₂Se thin film

Muscovite (K{Al₂[AlSi₃O₁₀](OH)₂}) mica, which is a widely used substrate for the growth of 2D materials thanks to its atomically flat surface at centimeter scale, was chosen as the substrate for the growth of uniform Bi₂O₂Se thin film. As shown in Figure S1, the contact angle between muscovite and Bi(NO₃)₃/(CH₂OH)₂ solution was as small as 6° (Figure S1), which suggested excellent affinity between muscovite and the solution, confirming the Bi-containing precursor superbly uniform on the muscovite.



Figure S1. Contact angle test of muscovite mica and Bi(NO₃)₃/(CH₂OH)₂.

Bi(NO₃)₃·5H₂O was chosen as the precursor of Bi₂O₃ thin film, because it can decompose into Bi₂O₃ at relatively low temperature without any residue. The reaction proceeded as follows: Bi(NO₃)₃·5H₂O → Bi₂O₃ + NO_x + O₂ + H₂O. According to Thermogravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC) tests (Figure S2), the difference between the mass of Bi(NO₃)₃·5H₂O and decomposed Bi(NO₃)₃·5H₂O is 51.9% derived from the DSC, which was identical to [2M(Bi(NO₃)₃·5H₂O) - M(Bi₂O₃)]/2M(Bi(NO₃)₃·5H₂O) (52%). It was indicated that Bi(NO₃)₃·5H₂O was decomposed completely into Bi₂O₃ at 529 °C. In our experiment, in order to obtain uniform Bi₂O₃ thin film, we set the decomposition temperature at 480 °C and annealed the Bi(NO₃)₃ thin film for a long time.

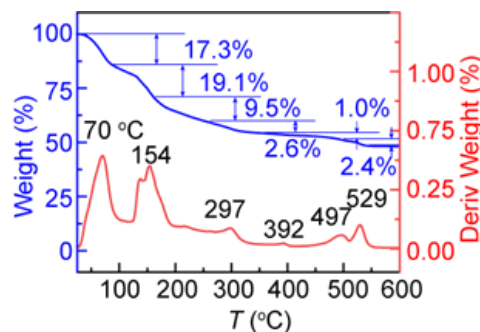


Figure S2. Thermogravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC) curves of Bi(NO₃)₃·5H₂O

2. Morphology and crystal structure of Bi₂O₃ thin film

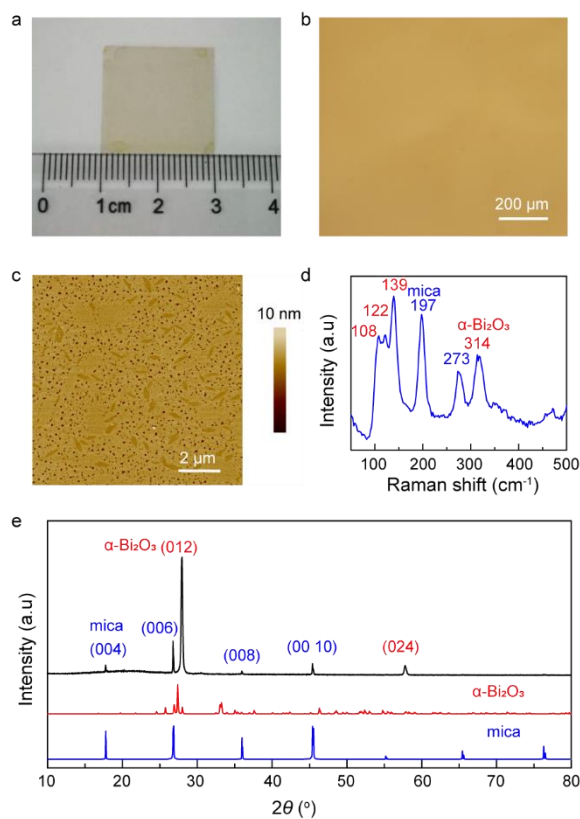


Figure S3. Morphology and structure analysis of Bi_2O_3 thin film. (a) Photograph of Bi_2O_3 thin film; (b) Optical microscopy image of Bi_2O_3 thin film with excellent uniformity; (c) Atomic force microscopy (AFM) image of Bi_2O_3 thin film; (d) Raman spectrum of Bi_2O_3 thin film showed that the Bi_2O_3 is of α -phase; (e) XRD pattern of Bi_2O_3 thin film on mica verified the crystal structure and it is aligned to (012) crystal plane.

3. Thickness control of $\text{Bi}_2\text{O}_2\text{Se}$ thin film

The thickness of $\text{Bi}_2\text{O}_2\text{Se}$ thin film can be readily controlled by changing the rotate speed as spin-coating $\text{Bi}(\text{NO}_3)_3/(\text{CH}_2\text{OH})_2$ solution. The morphology of $\text{Bi}_2\text{O}_2\text{Se}$ thin films with different thickness was imaging by AFM, as shown in Figure S4. These $\text{Bi}_2\text{O}_2\text{Se}$ thin films were uniform and continuous.

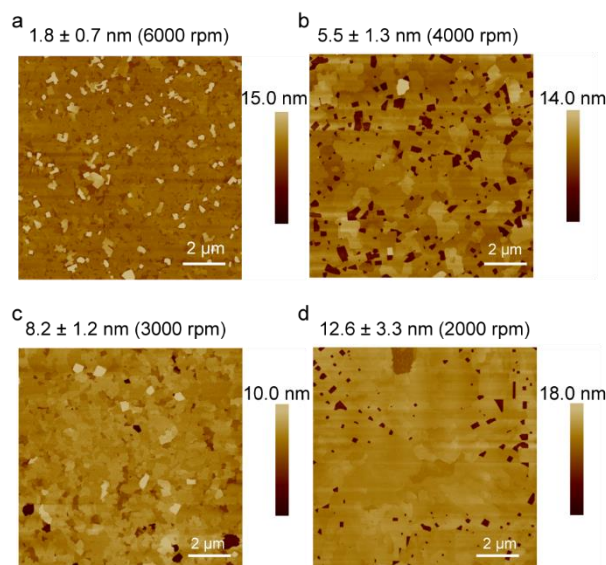


Figure S4. Morphology of $\text{Bi}_2\text{O}_2\text{Se}$ thin films with different thicknesses

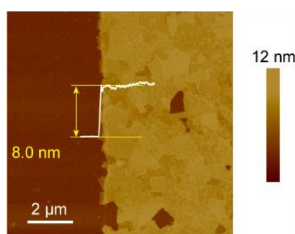


Figure S5. AFM image of $\text{Bi}_2\text{O}_2\text{Se}$ thin film prepared at 3000 rpm, which is etched into discrete sheets by wet chemical method.

4. Fabrication of devices on flexible substrate

A transfer technique was implemented after device fabrication. FETs were transferred onto flexible substrate polyvinyl chloride (PVC) with assist of thermal release tape. Firstly, thermal release tape was attached to the surface of FETs on muscovite (Figure S6a). Secondly, thanks to its layer structure, thick muscovite would be cleaved leaving thin muscovite with devices attached to the thermal release tape (Figure S6b). Then the thermal release tape/devices/thin muscovite structure could be stick to PVC with double-sided tape (Figure S6c). Finally, the above-mentioned structure would be heated at 100 °C to release the thermal release tape (Figure S6d). This strategy can be expanded to other materials growing on layered hard substrates for flexible electronics.

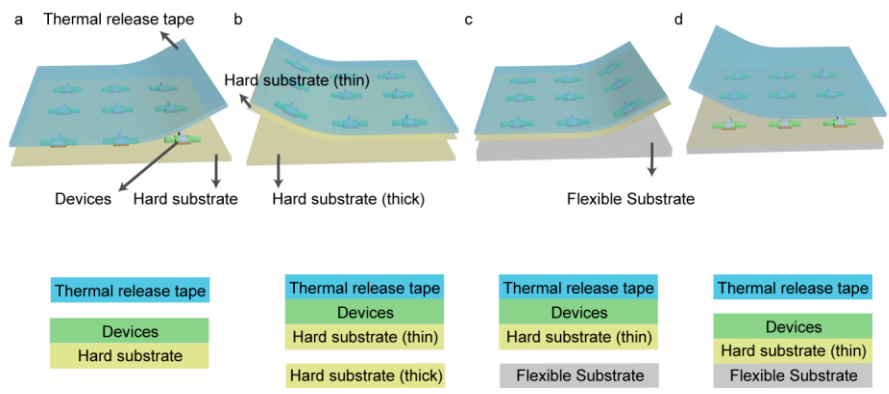


Figure S6. Scheme of devices transferred onto flexible substrate