Supporting information of

High-Mobility InSe Transistors: the Role of Surface Oxides

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Removing ultrathin $InSe_{1-x}O_x$ with argon plasma

Even for the as-exfoliated bulk InSe flakes, the XPS spectra of O 1S shows that there is still a noticeable peak, meaning the oxidation is considerably quick and inevitable. Owing to a very short oxidation period and intrinsic layer structure of InSe, the surface oxidation is expected to be $InSe_{1-x}O_x$. The $InSe_{1-x}O_x$ is ultrathin so that it can be removed by 3-second argon plasma etching. The O 1S peak completely disappeared after plasma etching as shown in Figure S1. Similarly, the dry-oxidation treatment forms an ultrathin $InSe_{1-x}O_x$ on the top of InSe. It can quickly be removed by argon plasma etching, too.

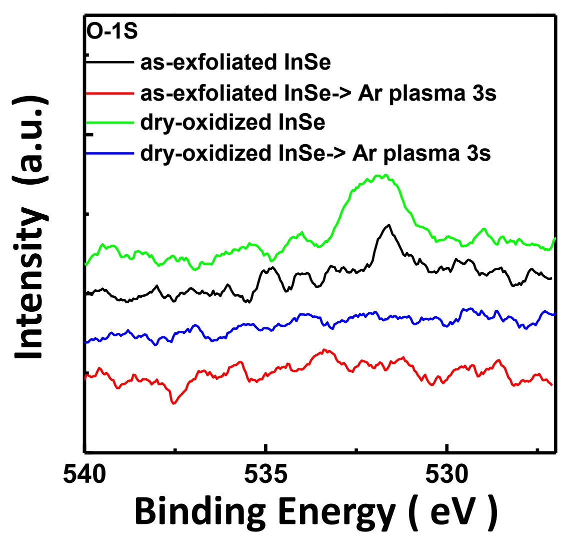

Figure S1 O 1S spectra of as-exfoliated and dry-oxidized InSe before and after etched by argon plasma.

Oxidation products of InSe

Figure S2 is the In $3d_{5/2}$ peak for air-oxidized InSe. The broad peak can be deconvoluted into three subpeaks which stand for In_2O_3, In_2Se_3, and InSe, respectively.

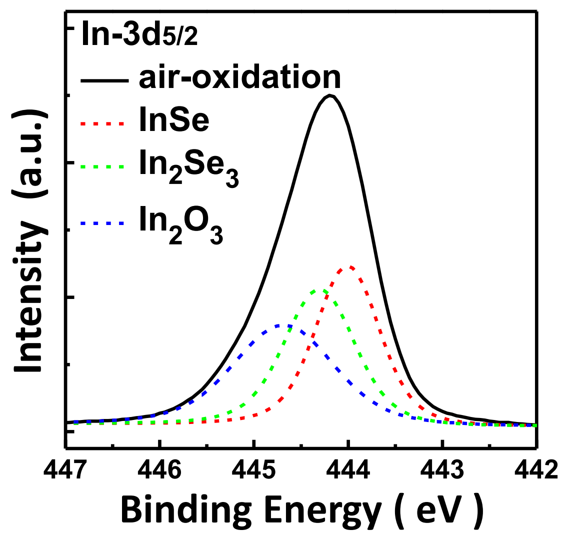

Figure S2 In $3d_{5/2}$ spectrum of air-oxidized InSe.

Dry-FET fabricated by conventional e-beam lithography

For dry-FETs fabricated by conventional e-beam technique, the source-drain patterns were defined by e-beam lithography as shown in Figure S3a. Because the e-beam technique includes process of heating resist and exposure to high energy electron beam, the surface oxide layer is expected to be thick under such condition. Therefore, the gate-dependent conductivity in Figure S3b shows a strong hysteresis phenomenon with a low conductivity. This strong hysteresis would simultaneous cause an unstable current and a fast current saturation as shown in Figure S3c.

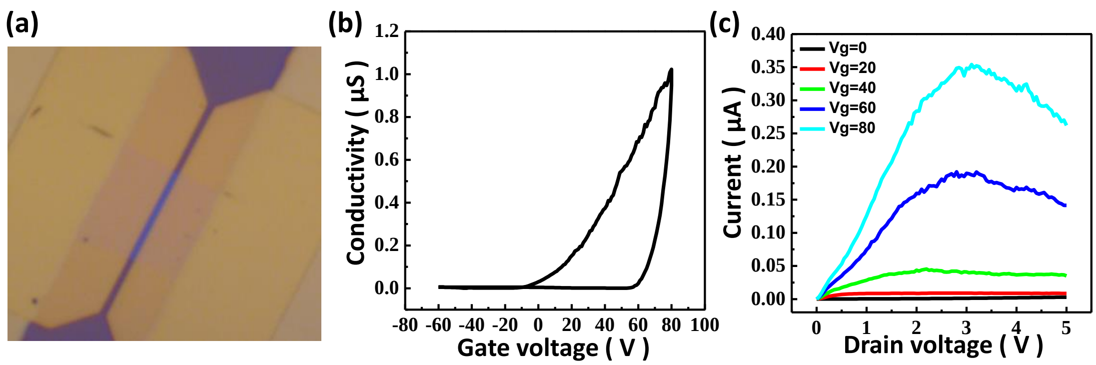

Figure S3 Dry-FET fabricated by e-beam lithography. (a) Optical image of the device fabricated by e-beam. (b) Gate-dependent conductivity and (c) I-V_d characteristic of the dry-FET fabricated by e-beam lithography.

Metal-insulator transition (MIT) of InSe

Figure S4 is the room-in diagram of gate-dependent conductivity shown in Figure 4d. The conductivity in y-axis is transferred to quantum conductivity (e^2/h) as shown in the figure. The MIT occurs at the range of 1-3 quantum conductivity as marked in the figure.

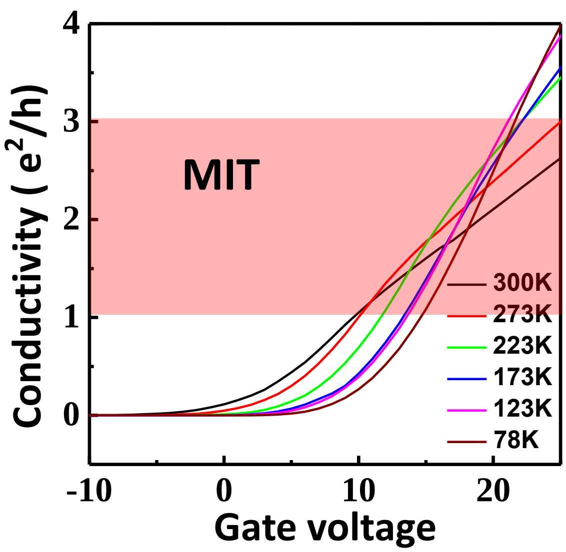

Figure S4 Metal-insulator transition of a dry-oxidation InSe device.

Device with as-exfoliated InSe

An as-exfoliated InSe flake without oxidation pretreatment was used to fabricate transistors. Figure S5a shows the gate-dependent conductivity of such device. A large hysteresis is observed in the diagram, indicating there is still a obvious oxidation effect for this device. Because the device was fabricated in air, the spontaneous oxidation was inevitable. The oxide layer results in large hysteresis and obvious fast current saturation as shown in Figure S5a and b.

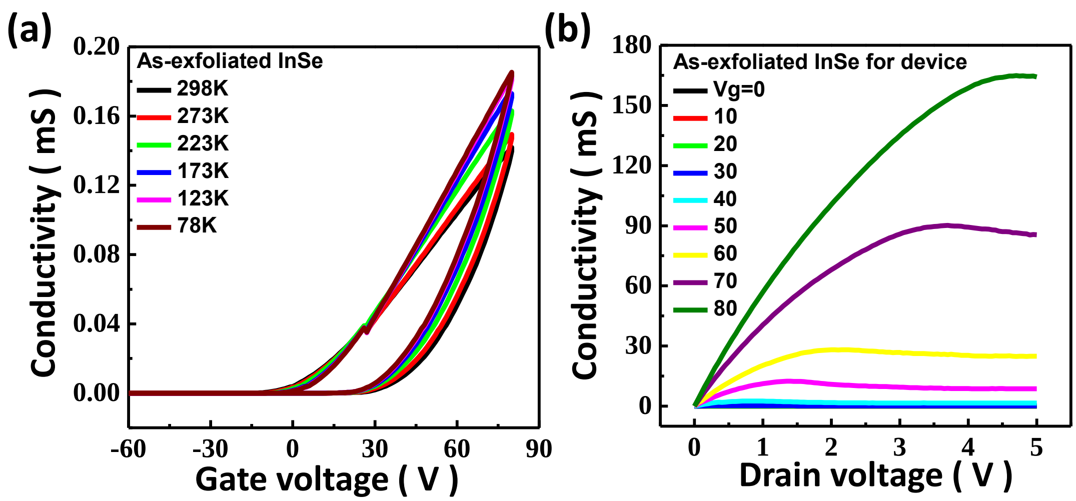

Figure S5 Transport characteristic of as-exfoliated InSe without oxidation pretreatment. Gate-dependent conductivity and I-V_d characteristic are shown in a and b, respectively.

Stability of dry-FET

The stability of dry-FET was tested under two kinds of conditions. The device was first put in a pure oxygen environment for 4 days. The transport property shown in Figure S6 exhibits that the performance does not obvious change under this condition, meaning that the dry-oxidation process is self-limited atop of InSe. However, as placed in air, the water could still slowly damage the dry oxide layer by its strong H-bond,[1] resulting in further oxidation. Therefore, the conductivity decreased accompanied with a larger hysteresis after placed in air for one month as shown in the Figure S6.

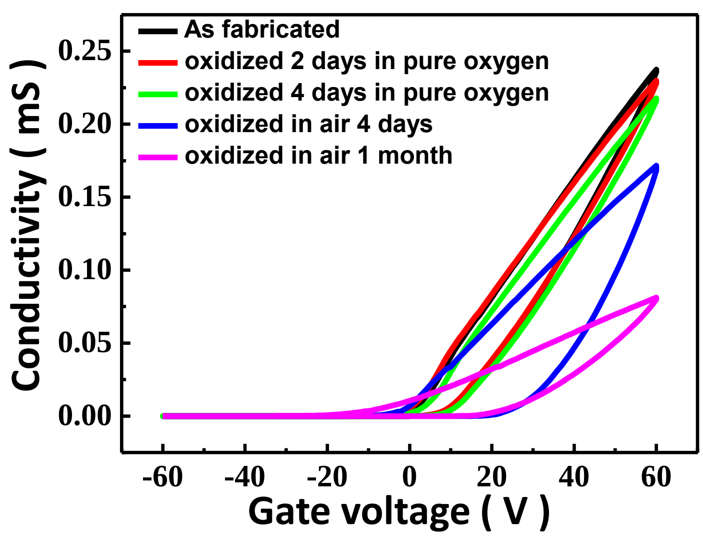

Figure S6 Stability test of the InSe device with dry oxide.

Light-assisted oxidation process of InSe

The formation of superoxide anions atop of InSe is the first step for oxidation as described in the manuscript. This process occurs when excitons donate their electrons to oxygen molecules. Therefore, the formation of superoxide anions would depend on the existence of light. Figure S7 compares the O 1S spectra of InSe oxidized under dark and light conditions in air. The sample oxidized under light illumination shows a higher O 1S signal, certifying the light-induced mechanism.

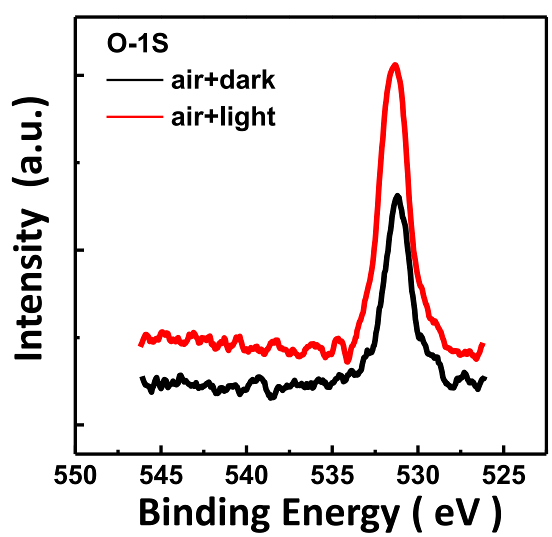

Figure S7 O 1S spectra of InSe oxidized under dark and light environments.

Dry-FET with ultrathin InSe (4nm)

A 4nm InSe flake was used to fabricate dry-FET. This device can not be turned on even applying a 130V back-gate bias as shown in Figure S8, which indicates the *p*-type doping effect of the oxide layer is considerably strong. This result further confirms the thickness-dependent oxidation and doping effect described in the manuscript.

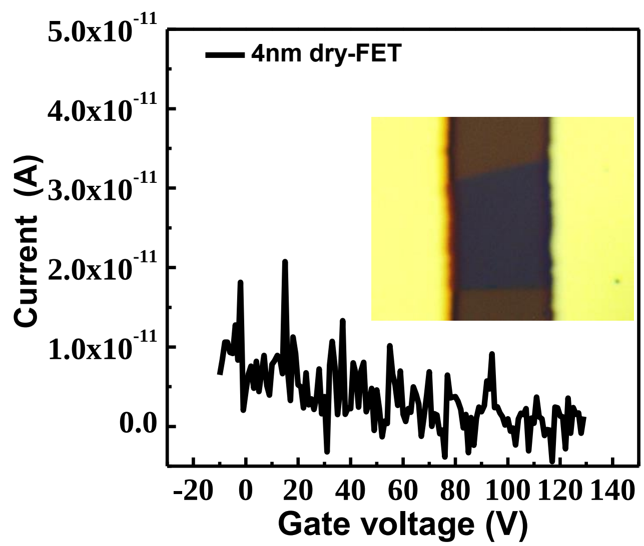

Figure S8 Transfer characteristic and optical image (inset) of a 4nm dry-FET.

REFERENCES

1. Zhou, Q.; Chen, Q.; Tong, Y.; Wang, J., Light- Induced Ambient Degradation of Few- Layer Black Phosphorus: Mechanism and Protection. *Angew. Chem.* **2016,** *128*, 11609-11613.