Schottky Barrier Reduction on Optoelectronic Responses in Heavy Ion Irradiated WSe$_2$ Memtransistors

Shengxia Zhang$^{1,2,*}$, Lijun Xu$^1$, Shifan Gao$^{1,3}$, Peipei Hu$^1$, Jiande Liu$^1$, Jian Zeng$^1$, Zongzhen Li$^1$, Pengfei Zhai$^1$, Li Liu$^1$, Li Cai$^1$, Jie Liu$^{1,*}$

$^1$ Materials Research Center, Institute of Modern Physics, Chinese Academy of Sciences (CAS), Lanzhou, 730000, P. R. China
$^2$ University of Chinese Academy of Sciences, Beijing, 100049, P. R. China
$^3$ Northwest Normal University, Lanzhou, 730070, P. R. China

1. Raman spectra of the pristine and irradiated samples

We found a number of features below and above the first-order mode in 5L-WSe$_2$ prepared by mechanical exfoliation method, as shown in Fig.S1. The two features located at 246.8 and 255 cm$^{-1}$ might correspond, respectively, to the first order $E_{2g}^1$ and $A_{1g}$ modes. The peaks at 116 cm$^{-1}$, 137 cm$^{-1}$, 352 cm$^{-1}$, 370 cm$^{-1}$, and 392 cm$^{-1}$ have been assigned as second-order Raman modes involving phonons at the M point. For most monolayer TMDC, $B_{12g}$ is the optically inactive mode. Its origin has been recently reported as an acoustic (A)-symmetry first-order mode corresponding to an interlayer vibration, which can be used to distinguish monolayer and multilayer WSe$_2$.

* Corresponding author. Tel: +86 931 4585250. Email: zhangsx@impcas.ac.cn (S. X. Zhang).
* Corresponding author. Tel: +86 931 4969334. Email: j.liu@impcas.ac.cn (J. Liu)
Fig. S1 Demonstration of the Raman peaks of the pristine and irradiated WSe$_2$ devices.

2. Microstructure of the tracks

Fig. S2 (a) Averagely distributed tracks induced by Ta ion irradiation, The
applied fluence is 5×10$^{10}$ ions/cm$^2$. (b),(c) TEM images of the carbon-film
supported few-layer WSe$_2$ film irradiated by Ta ions with an angle of 90° and
45°. The applied fluence is 5×10$^{10}$ ions/cm$^2$. (d) TEM images of the free
standing WSe$_2$ film irradiated by Ta ions with angles of 45°. The applied
fluence was 5×10$^{10}$ ions/cm$^2$.

3. I-V scanning loops of the pristine devices

Fig. S3 depicted the 20-turn I-V curve characteristics of the pristine devices under
electrical signal stimulation with light excitation ($P_{in}$=100 mW/cm$^2$). The I-V curves of
the pristine devices showed typical I-V characteristics without RS behaviors.
Particularly, the value of current under dark conditions was slightly greater than that
under light excitation.
Fig. S3 I-V scanning loops of the pristine devices with and without illuminance. The backgate voltage is kept as 20 V.

4. I-V scanning loops of the irradiated devices

Fig. S4 I-V scanning loops of the WSe\textsubscript{2}-based devices irradiates by SHIs with angles of 90° and 75°. The backgate voltage is kept as 20 V.

5. Simulation details

Self-consistent periodic DFT calculations were carried out using the Vienna ab initio simulation package ‘VASP’. Briefly, the exchange-correlation effects were treated within the generalized gradient approximation using the PBE functional. The ionic cores were described using the projected augmented wave (PAW) method, and the Kohn-Sham one-electron valence states were expanded in a basis of plane waves. Simplify processes of melting and quenching were setup to simulate the density of states (DOS) of heterostructure Cr (3 layers)/ WSe\textsubscript{2} (3 layers). The molten structure was quenched at a rate of 10\textsuperscript{14} K/s, which was in a similar timescale according to the spike model. Complex defects were introduced in the system. The spin-up and spin-down total DOS shown in Fig. S5(a) and (b)
were symmetric, which indicated that these systems exhibit the non-magnetic ground states. An obviously reduced DOS at $E_F$ was depicted for Cr in the defective heterostructure, resulting from strong chemical interactions between the metal electrodes and WSe$_2$ flakes, as seen in Fig.S5(c). Considering the computation and time consuming, the heterostructure model of Cr (3 layers)/ WSe$_2$ (3 layers) including 672 atoms were setup in this work. Combined with actual experimental conditions, high-energy heavy ions can completely penetrate 5 layers or 3 layers of WSe$_2$. Therefore, the conclusion related with irradiation induced interfacial mixing will not be affected when the model is smaller to 3 layers WSe$_2$.

Fig.S5 (a) Total DOS patterns of the pristine heterostructure Cr/WSe$_2$. (b) Total DOS patterns of the defective heterostructure Cr/WSe$_2$. (c) Comparison of the partial DOS of Cr $d$ states in the perfect and defective heterostructure.

6. **Sweep loops of the transfer characteristic of SHI irradiated devices**
Fig. S6 The sweep loops of the transfer characteristic of SHI irradiated devices. The transfer characteristics were measured under dark and illumination with power density of 100 mW/cm², which were labelled by hollow and solid circles in the figures. The swept directions were marked by red and blue arrows.

7. I-T curves of the devices irradiated with angles of 90° and 75°

Fig. S7 I-T curves of the devices irradiated with an angle of 90° and 75° under 630 nm pulses with intensity of 100 mW/cm². The width and intervals were increased from 3 s to 5 s. The applied voltage $V_{ds}$ was 5 V at $V_g = 0$ V.