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Synthesis of $NbSe_2$ Single-crystalline Nanosheet Arrays for UV Photodetectors

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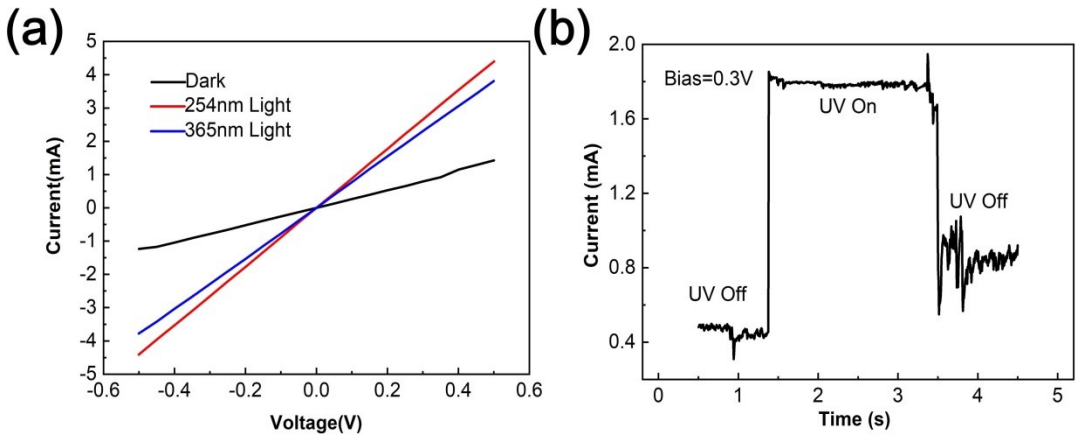

Figure S1. (c) The I-V curve of the $NbSe_2$ device is under dark, 254nm and 365nm laser irradiation. (d) The I-T curve of the $NbSe_2$ device at 0.3V bias under the irradiation of a 254nm laser.

Fig S1 shows the *I-V* and *I-T* characteristics of the NbSe2 nanosheet photodetector. Linear *I-V* indicates good contact between $NbSe_2$ nanosheets and electrodes. The Fig S1a clearly shows that when a device is measured in darkness, 356 nm and 254 nm UV light irradiation, the current increases gradually. The resistivity of the $NbSe_2$ nanosheet arrays is estimated to be 2.5 x 10^{-3} Ω·m under dark condition and while 7.5 x 10^{-4} Ω·m and 6.7 x 10^{-4} Ω·m under 365nm and 254nm UV light irradiation, respectively. Fig S1b shows the *I-T* (current-time) curve of the $NbSe_2$ device under 254 nm UV light irradiation. When the bias reaches 0.3V, the measured current is 1.8 and 0.4 mA, respectively. The on/off ratio of this device is 4.5, and the response time for both rise and fall is ~7 ms. The fast response to UV light of the devices is due to the high crystal quality of $NbSe_2$ nanosheets.

The denstiy fundtional theory (DFT) computations were performed using the projector augmented wave (PAW) method and Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation fuctional[1]. Monkhorst-Pack k-point grid of 8 ×8

×1 was used for structural relaxation, and 16 ×16 ×1 for self-consistent calculations. The convergence threshold was 10^{-4} eV in energy and 10^{-3} eV/Å in force. The cutoff energy used for the plane-wave basis set is 400 eV.

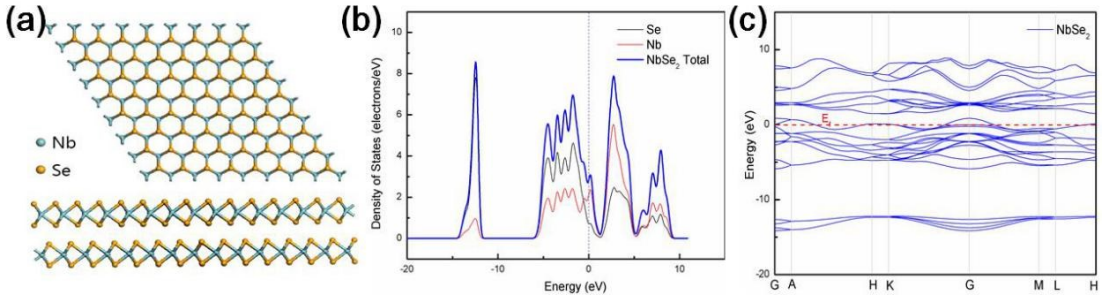

Figure S2. (a) Hexagonal crystal structure of 2H-$NbSe_2$, The blue and yellow balls represent Nb and Se atomic columns. (b) Using GGA-PBE to approximately estimate the state density of 2H-$NbSe_2$. (c) The energy band structure of $NbSe_2$ along the brillouin belt in the direction of high symmetry.

To further understand the microstructure and electrical properties of the $NbSe_2$ nanosheet arrays, we employed density functional theory (DFT) computation on 2H-$NbSe_2$. Fig S2(a) shows the overall structure of $NbSe_2$, which belongs to the hexagonal system with space group P-6m2. Each Nb atom is surrounded by six Se atoms, and has triangular prism symmetry [2, 3]. The stacking along the c axis starts from Se-Nb-Se, and a slab is formed in this order, with a strong covalent bond between the three atomic layers and a one-weak bond between the vdws-type slabs. Fig S2(b) shows the atom projected density of states (DOS) diagram for $NbSe_2$. It obviously shows that there is a partial wave DOS across the Fermi level, which confirms the semimetal nature of the 2H- $NbSe_2$. The DOS below the Fermi level is dominated by the Se element and energy above the Fermi level is dominated by the

Nb element[1]. The energy band structure of 2H-$NbSe_2$ along the highly symmetric direction is shown in Fig S2(c), where the GGA-PBE approximation is used in the calculation. As shown in Fig S2(c), the compound exhibits a special energy band structure, which represents a layered structural system，in which the intermediate band located in energy gap around E_f. The presence of bands crossing the E_f means that 2H-$NbSe_2$ is in semimetic state, and these bands originate mainly from Se 4p and partial from Nb 4d states, indicating that there is a hybrid between them[2]. The calculated band structure of 2H-$NbSe_2$ is validated by the experimental data from angle-resolved light emission spectra (ARPES)[4].

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