Supplementary Information (SI) for Materials Chemistry Frontiers.

Supporting Information for

Dopant-Mediated Carrier Tunneling in Short-Channel Two-Dimensional Transistors

Yue Lu[1#], Chenyu Li[1#], Shenbo Yang[2], Mingxuan Yuan[1], Shuo Qiao[1], Qingqing Ji[1*]

[1]School of Physical Science and Technology, ShanghaiTech University, Shanghai 201210, China

[2]Hongzhiwei Technology (Shanghai) Co., Ltd., Shanghai 201210, China

[#]These authors contribute equally to this work.

*To whom correspondence should be addressed. E-mail: jiqq@shanghaitech.edu.cn

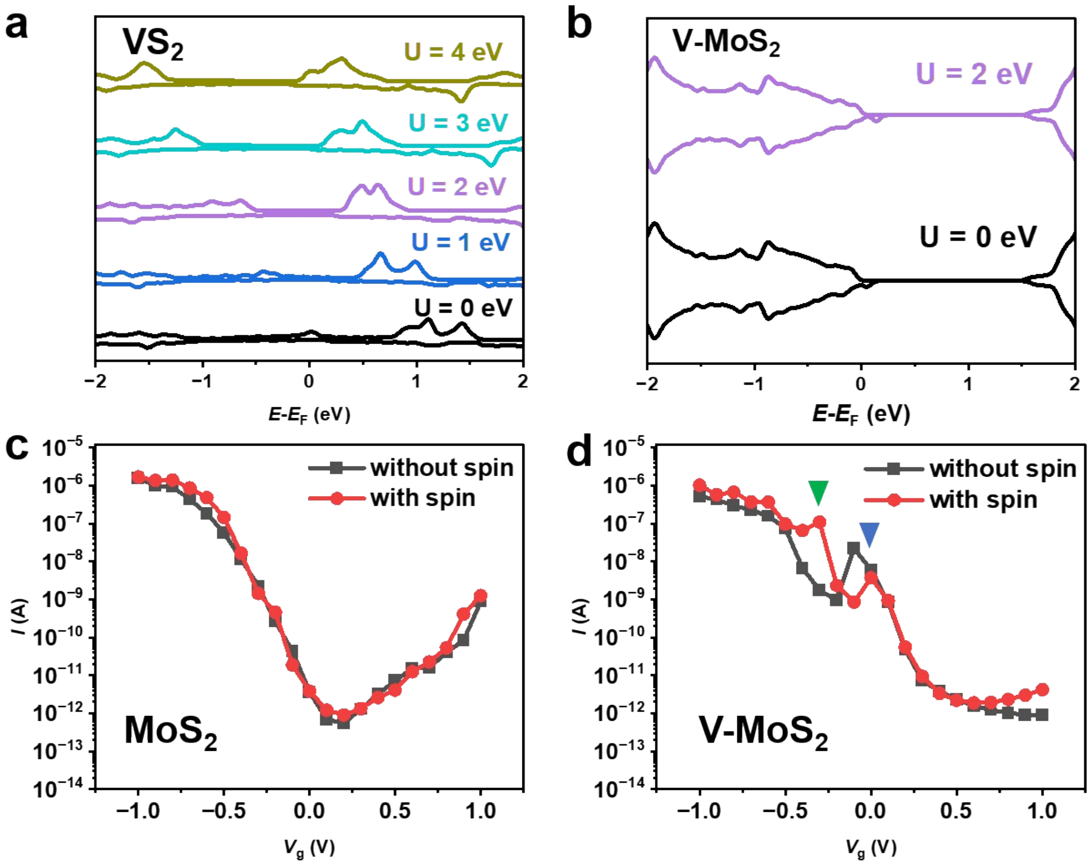

Figure S1. Magnetism in VS_2 and V-MoS_2 and its impact on transport. DOS of VS_2 (a) and V-MoS_2 (b) under different U values. Transfer characteristics of MoS_2 (c) and V-MoS_2 (d) with and without spin consideration at L_{ch} = 3 nm.

The impact of magnetism on the transport characteristics was studied. First, the density of states (DOS) of VS_2 and V-MoS_2 under different U values was calculated.

As shown in **Fig. S1a,b**, the U value only altered the band edge positions of VS_2 without affecting its metallic nature, and had negligible impact on V-MoS_2, where the V-doped state persists to be spin polarized. Then, a model system with a channel length of 3 nm was used to calculate the transfer characteristics considering magnetism. As shown in **Fig. S1c**, since the MoS_2 has no inherent magnetism, the two curves with and without considering spin overlap almost entirely. However, regarding the transfer characteristics of V-MoS_2, we can see that when magnetism is considered, the transconductance peak in the subthreshold region (as shown in **Fig. 3d** of the main text) splits into two kinks marked by blue and green arrows in **Fig. S1d**. This phenomenon possibly originates from the Coulomb splitting effect where the interaction of spin-up and spin-down electrons strongly depends on the spatial extension of the localized doping state (*Science* **2016**, *352*, 437-441). Importantly, it should be noted that although magnetism does affect the transport behavior of V-MoS_2, this does not change our key conclusion that the V-doped state can induce the assisted-tunneling effect under short-channel conditions.

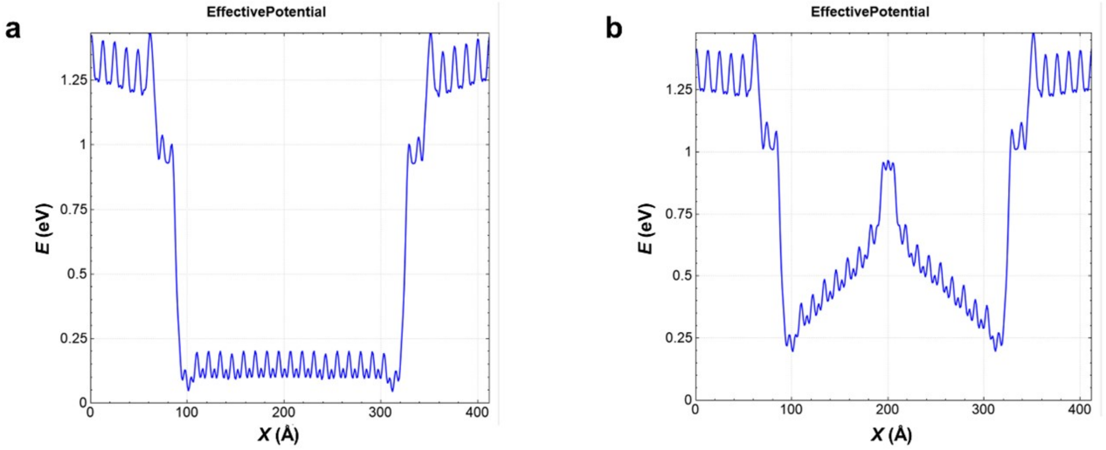

Figure S2. The effective potential of VS_2-MoS_2-VS_2 (a) and VS_2-V doped MoS_2-VS_2 (b) device.

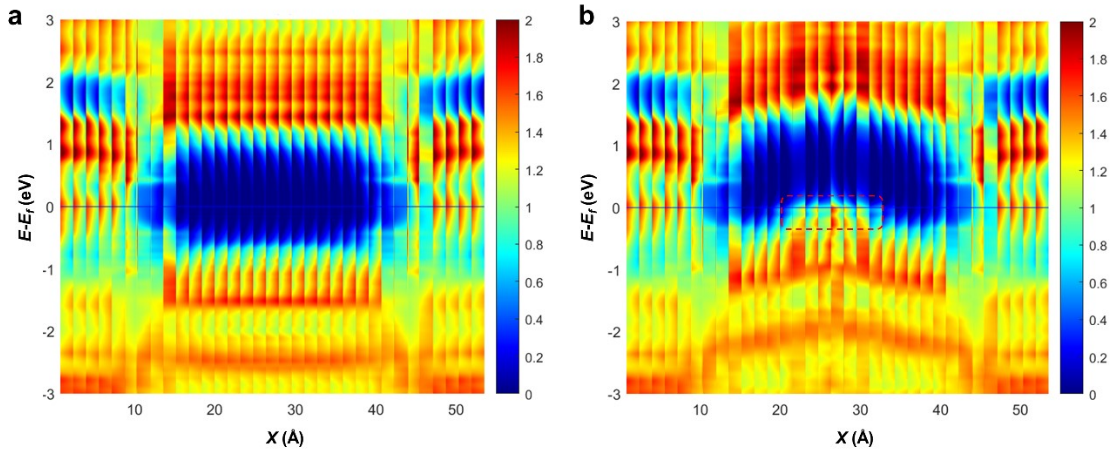

Figure S3. PDOS of VS$_2$-MoS$_2$-VS$_2$ (a) and VS$_2$-V doped MoS$_2$-VS$_2$ (b) device without bias and gate voltage.

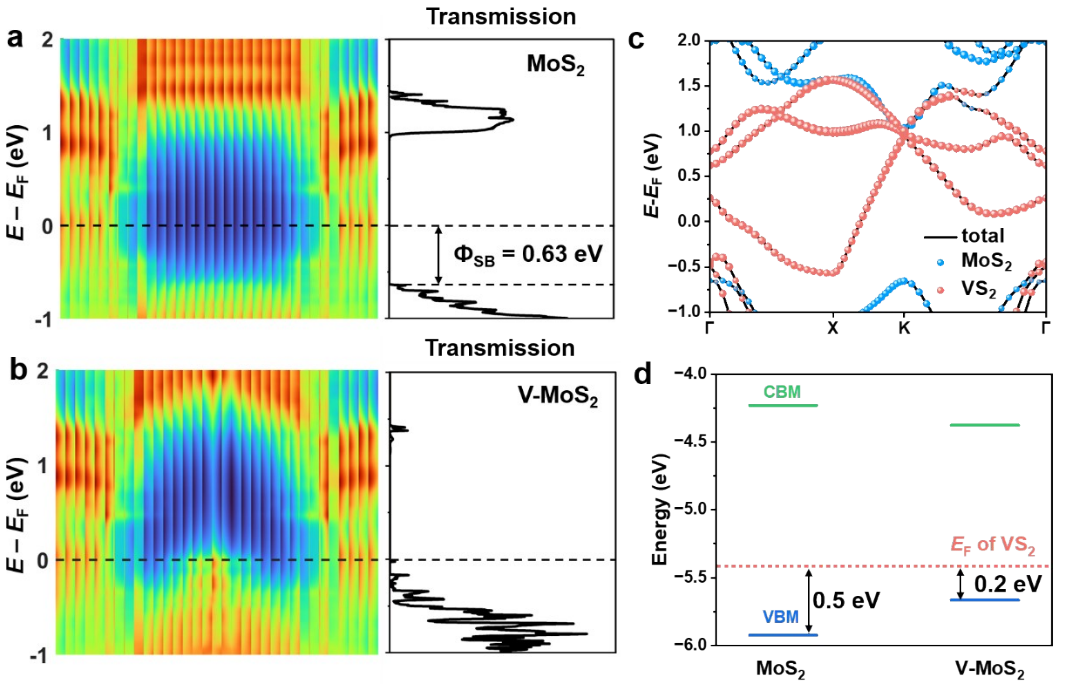

Figure S4. PDOS and electron transmission spectra for the VS$_2$-MoS$_2$-VS$_2$ (a) and VS$_2$-V doped MoS$_2$-VS$_2$ (b) at equilibrium with $L_{\mathrm{ch}} = 3$ nm. (c) Projected band structure of the VS$_2$-MoS$_2$ out-plane heterojunction. (d) The band edges of the two materials and the Fermi level of VS$_2$ relative to the vacuum level.

To elucidate the contact types, the projected density of states (PDOS) and electronic transmission spectrum of the device at equilibrium with a channel length $L_{\mathrm{ch}} = 3$ nm was first calculated. As shown in **Fig. S4a**, when the channel material is pristine MoS$_2$, a p-type Schottky contact is formed. From the transmission spectrum, the

Schottky barrier height for holes is determined to be 0.63 eV. When the channel material is V-MoS_2, as depicted in **Fig. S4b**, the Schottky barrier at this channel length is almost negligible due to the assisted-tunneling effect of V atoms, manifesting as a quasi-Ohmic contact. Furthermore, we also computed the contact properties of vertical VS_2/MoS_2 heterojunctions using the DS-PAW software. As illustrated in **Fig. S4c,d**, a p-type Schottky contact is also formed between MoS_2 and VS_2, with a Schottky barrier height of 0.5 eV (this value differs from the in-plane Schottky barrier height due to the different functionals used for Nanodcal and DS-PAW). Besides, for the vertical VS_2/V-MoS_2 heterojunction, a significantly lowered hole Schottky barrier of 0.2 eV is revealed, highlighting the vital role of V-doped states in modifying the interface properties.

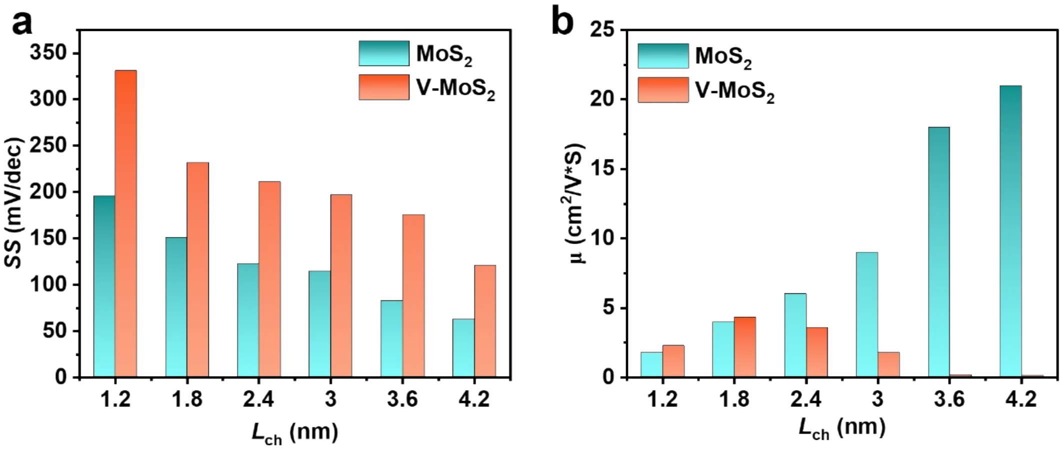

Figure S5. Subthreshold swing (a) and carrier mobility (b) at different channel lengths.

As observed in **Fig. S5a**, the subthreshold swing (SS) exhibits an increasing trend with decreasing channel length, which is a typical characteristic of the short-channel effect. Furthermore, as shown in **Fig. S5b**, when the channel length is less than 1.8 nm, the carrier mobility of the V-MoS_2 is slightly higher than that of MoS_2, owing to the assisted tunneling effect induced by the V dopants. Conversely, when the channel length exceeds 2.4 nm, the assisted tunneling effect is suppressed in the on-state, and the impurity scattering becomes the dominant mechanism, leading to a lower carrier mobility than that of MoS_2.

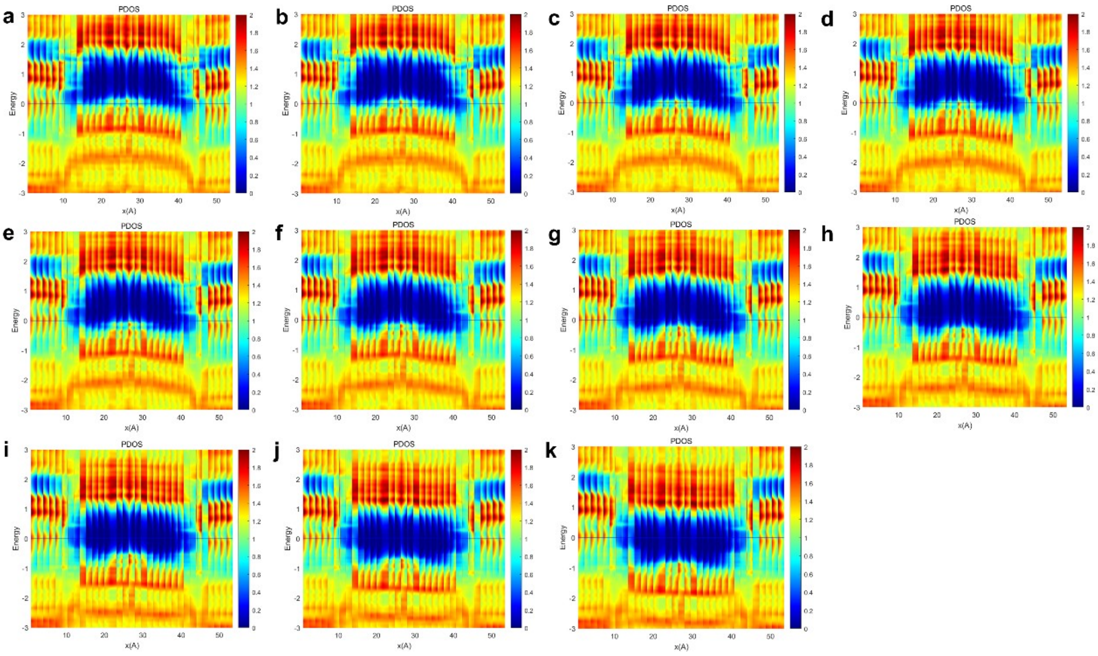

Figure S6. PDOS of VS_2-V doped MoS_2-VS_2 device under the gate voltage varying from 1 V to -1 V (a-k).

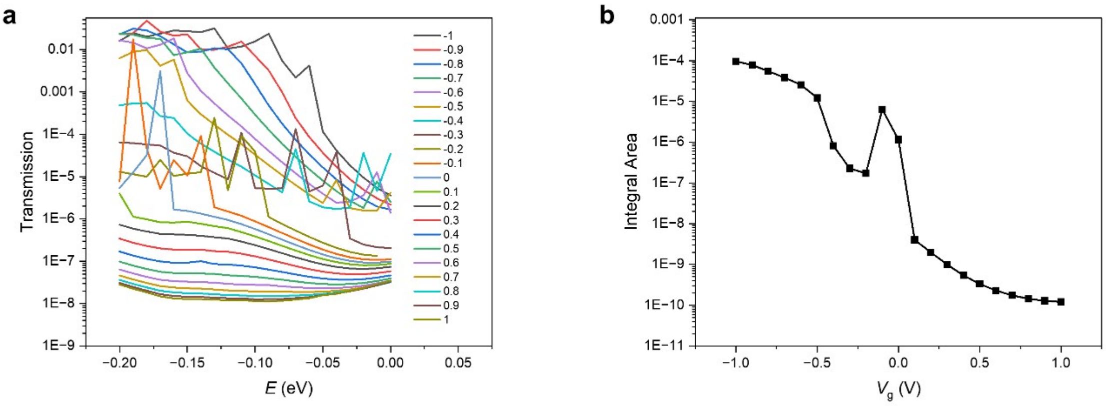

Figure S7. The transmission coefficient for gate voltage ranging from -1 V to 1 V (a) and corresponding integral area (b).