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Supporting Information for

Charge Transfer in Crystalline Germanium/Monolayer MoS2 Heterostructure Prepared by Chemical Vapor Deposition

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Characterization of monolayer MoS_2 The MoS2 samples were characterized before Ge thin film growth by Raman and photoluminescence (PL) spectroscopy. Figure 1 shows the room-temperature Raman and PL spectra of MoS_2 grown on a 150 nm-thick SiO_2/Si substrate. The peaks of Raman and PL spectra correspond to the features of monolayer MoS_2.[1,2]

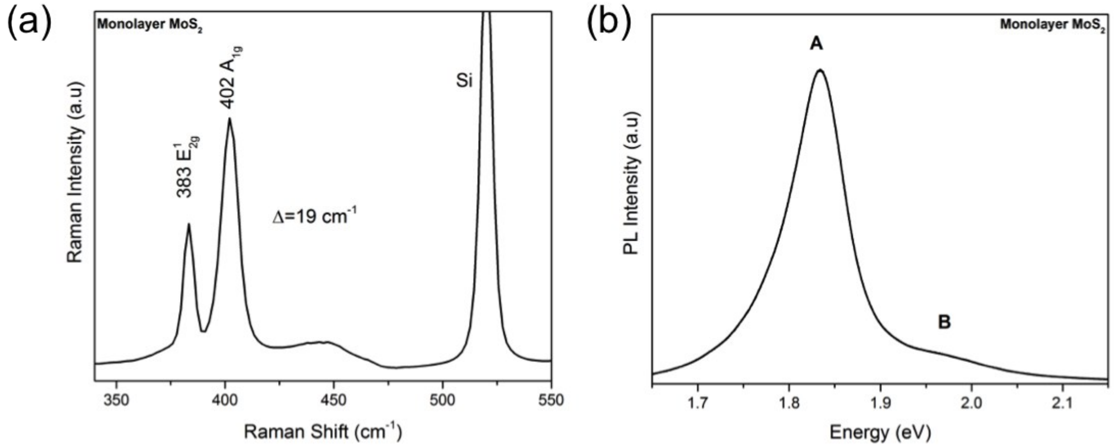
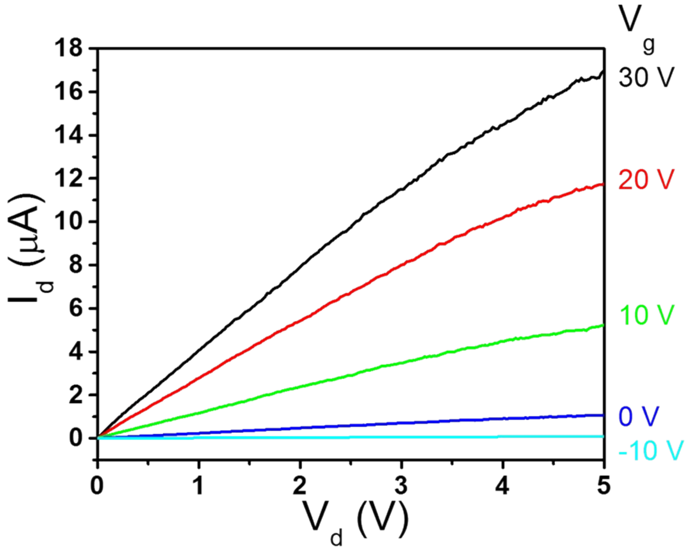


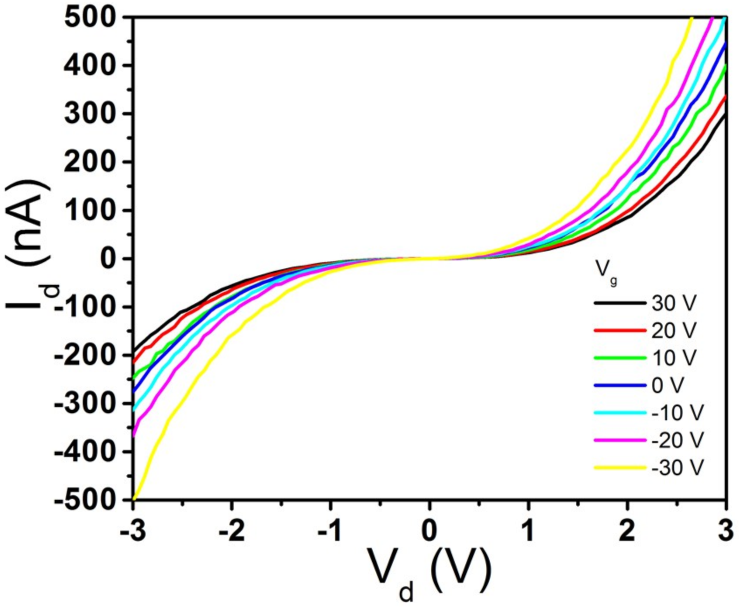
Figure S1. Room-temperature Raman (a) and photoluminescence (b) spectra of monolayer MoS_2 for Ge growth.

Figure S2. *I_d–V_d characteristic curves of MoS_2 before and after Ge growth (corresponding to Figure 4(a) and 4(b) in the main manuscript*

[Before Ge growth]

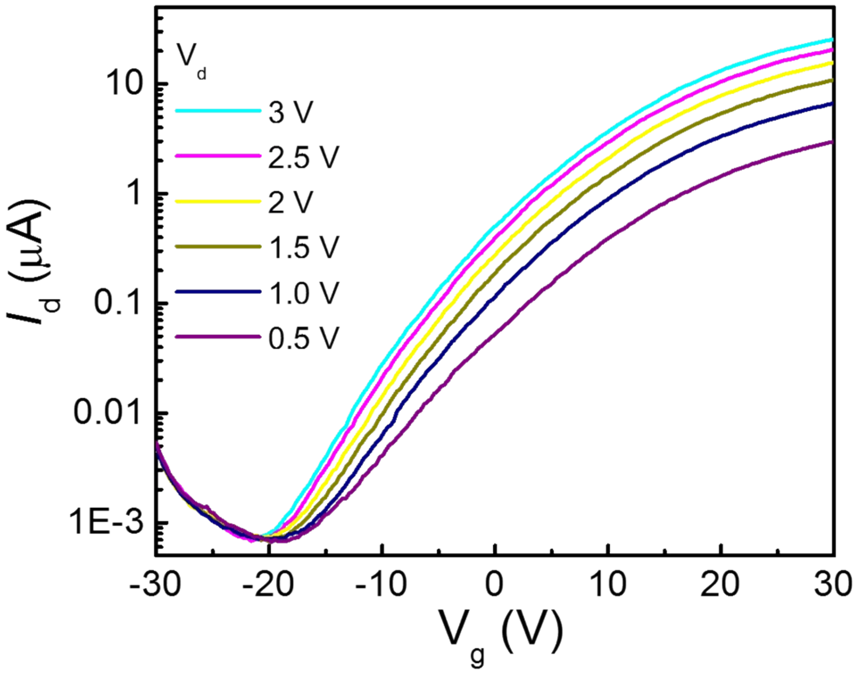


[After Ge growth]



The non-ohmic behavior observed in the I_d–V_d characteristics curve of MoS_2 after Ge growth is presumably related to the location of electrodes on MoS_2. The electrodes on MoS_2 and some region of MoS_2 nearby the electrodes are not covered with Ge as shown in Figure 2. Thus, the circuit of MoS_2 after Ge is equivalent to two positive channel metal oxide semiconductors in series.

Figure S3. *I_d–V_g characteristic curves of monolayer MoS_2 without (top) and with (bottom) Ge layer: Log plots of Figure 4(a) and (b)*



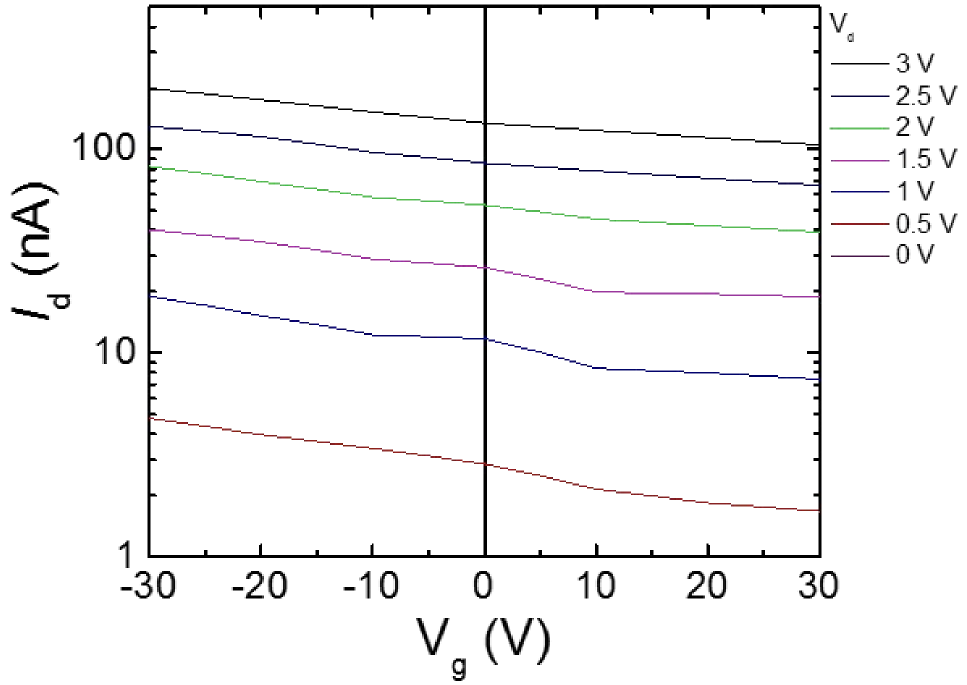


Figure S4. *I_d–V_d characteristic curve of Ge thin film grown on monolayer MoS_2 with top-gate configuration*

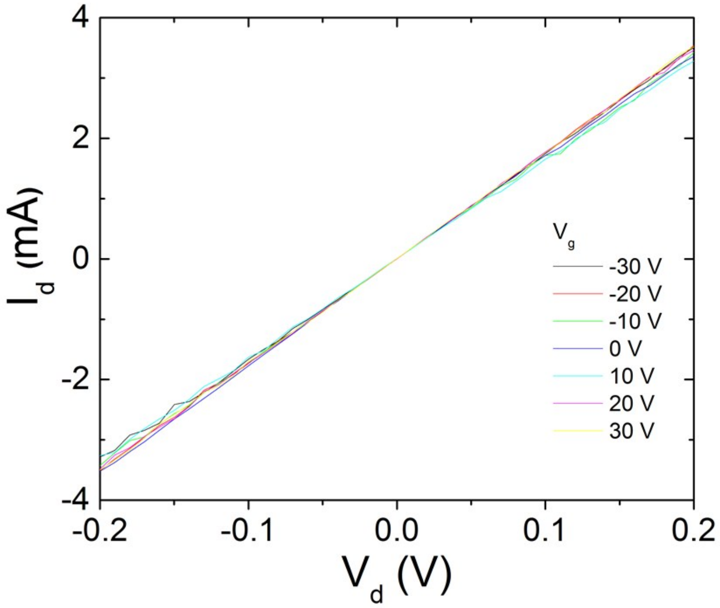
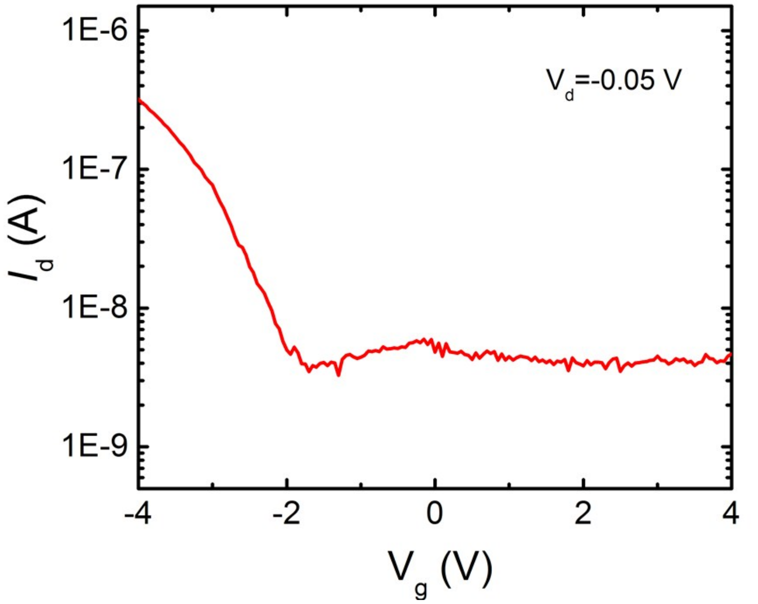


Figure S5. *I_d−V_g characteristic curve of Ge thin film grown a SiGe-buffer/intrinsic Si substrate*



Density Functional Theory (DFT) Calculations In this report, we theoretically demonstrate the possibility of self-doping between two semiconducting materials Ge and monolayer MoS_2. Our DFT calculations with Perdew, Burke, and Ernzerhof (PBE) exchange-correlation functional detect the presence of small band gap in bulk Ge, and about 1.6 eV band gap in monolayer MoS_2. Both band gaps are slightly under-estimated due to the well-known limitation of ground state nature of DFT, and can be treated using quasi-particle correction (e.g. GW[3,4]). But for the purpose of the current discus- sion, such treatments are not essential as long as our DFT calculations correctly determine the presence of semiconducting band gap. This also confirms the validity of our calcuations in determining the semiconducting or metallic nature of a given system. Both bulk Ge and monolayer MoS_2 is observed to have semi-conducting band gap in our first-principles calculation using DFT. We combined double layer Ge (110) crystal, and single layer MoS_2 to form Ge/MoS_2 heterostructure. Our calculation suggests the combined system to turn metallic due to possible charge transfer between the layers. Such self-doping effect has also been observed in recent experiment by one of the co-authors.

All calculations in this paper are performed using the plane-wave pseudo-potential code VASP[5,6] under the generalized gradient approximation of PBE.[7] For atomic core-levels, we have used projected augmented wave (PAW) potentials[8,9] treating the $4s4p$ of Ge, $4p5s4d$ of Mo, and $3s3p$ of S as the explicit valence electrons. A maximum energy cutoff of 360 eV is used for plane-wave basis set.

In our calculations, the bulk Ge has cubic unit cell with diamond like structure (lattice const. $a = 5.658Å$). For monolayer MoS_2, we have considered a supercell twice the primitive unit cell for honeycomb structure with lattice constants $a = b = 6.338\,Å$, $c = 20Å$, $\alpha = \beta = 90^o$, and $\gamma = 120^o$.

Ge (110) surface has honeycomb structure. In our tetragonal supercell, we have considered 5×6 unit cells of MoS_2, and 6x4 unit cells of Ge in 110 direction. The lattice constants for the tetragonal supercell are $a = 16.7Å$, $b = 15.9Å$, and $c = 30.0Å$.

Finally, we have relaxed our Ge/MoS_2 heterostructure using van der Waals (vdW) interaction. To keep the calculation expense reasonable, we have only considered four layers of Ge {110} planes. But in reality, such Ge layers can be much thicker. To mimic the bulkness of Ge layer, we kept the Ge ions frozen while relaxed the Mo and S ions using vdW interaction.

To incorporate the vdW interaction between the graphene and MoS_2 layers, we have used optB86b-vdW functional where the exchange functionals were optimized for the correlation part.[10]PBE functional is removed by using the parameter AGGAC =0.000 in the input file in order to avoid double-counting.

For ionic relaxation of Ge/MoS_2 systems, we have used the Γ point to sample the Brillouin zone, while for all other calculations, e.g., SCF, and density of states (DOS), we used 7x7x1 k-points in the Brillouin zone. For bulk Ge, our self-consistent (SCF) calculation used 11x11x11 k-points where the DOS calculation involved 31×31×31 k-points in the Brillouin zone. For monolayer MoS_2, we used 11×11×1 k-points for SCF and 21×21×1 k-points for DOS calculations.

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