Modulation of Structural and Electronic Phases in Two-dimensional $PdSe_2$

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Tuning the ambipolar behaviour in charge carrier transport via defect-engineering is crucial for achieving high mobility transistors for nonlinear logic circuits. Here, we present the electric-field tunable electron and hole transport in a microchannel device consisting of highly air-stable van der Waals (vdW) noble metal dichalcogenide (NMDC), $PdSe_2$, as an active layer. Pristine bulk $PdSe_2$ offers ambipolar transfer characteristics with a slight electron dominance recorded in field-effect transistor (FET) characteristics showing an ON/OFF ratio < 10 and electron mobility ~21 $cm^2/(V.s)$. However, transfer characteristics of $PdSe_2$ can be tuned to a hole-dominated transport while using hydrochloric acid (HCl) as a p-type dopant. On the other hand, the chelating agent EDTA, being a strong electron donor, enhances the electron-dominance in $PdSe_2$ channel. In addition, p-type behaviour with a 100 times higher ON/OFF ratio is obtained while cooling the sample down to 10 K. μ-focused angle-resolved photoemission spectroscopy also resembles the p-type band structure of $PdSe_2$ single crystal at low temperature. In addition, the pressure-induced structural modulation associated with metallization were also investigated by high pressure X-ray diffraction and Raman spectroscopy study. $PdSe_2$ exhibits high in-plane auxeticity under compression. Around 4.7 GPa, it undergoes an orthorhombic to pyrite (cubic) structural transition with a significant reduction of the interlayer vdW gap resulting in a 3D type network. Raman spectroscopy and *ab initio* electronic structure calculations at the transition pressure regime reveal anomalous phonon mode softening with the weakening of Se-Se dimer bonds and interlayer charge transfer followed by a semiconducting to metallic transition. Also, around 8.8 GPa, a new marcasite phase arises that coexists with the cubic phase. Such an anisotropic and highly auxetic vdW architecture may open up new possibilities towards next-generation electronics as well as exotic superconducting phases under external perturbation.

References

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Figures

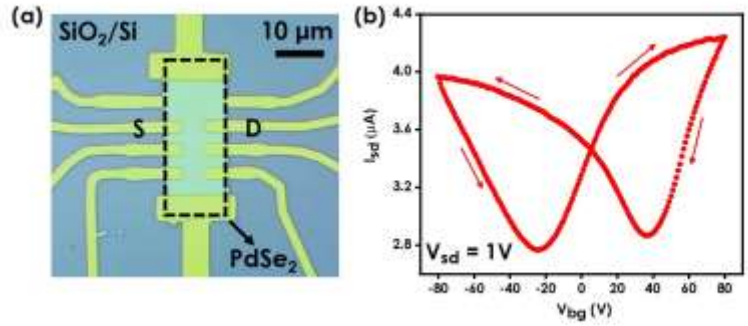

Figure 1: (a) Optical micrograph of a multilayered $PdSe_2$ field-effect transistor device. (b) Two-terminal ambipolar transfer characteristics at room temperature.