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**Supplementary Information**

Quantum confinement-induced semimetal-to-semiconductor evolution in large-area ultra-thin PtSe$_2$ films grown at 400 °C

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Supplementary Figure 1. Band structure and DoS of “bulk” PtSe$_2$ illustrating the overlap of valence and conduction bands.

Supplementary Figure 2. Top view (up panel) and side view (down panel) of geometric structures of pristine (left) and defective with Pt vacancy (right) PtSe$_2$ monolayer. Dark blue and orange atoms are Pt and Se atoms, respectively. $b$, $d$ and $\odot$ represent bond length, distance and Pt vacancy, respectively.
Supplementary Figure 3. Band structure of monolayer PtSe$_2$ supercell with Pt vacancy before unfolding.

Supplementary Figure 4. Partial density of states (PDOS) of nearest Se and nearest Pt atoms to Pt vacancy compared to PDOS of next nearest Pt atom, showing strong localized nature of the Pt vacancy in PtSe$_2$. 
Supplementary Figure 5. Cross-sectional TEM image of transferred (a) 2.5-3 nm, and (b) 5-6.5 nm PtSe$_2$ samples. Scale bar is 5nm.

Supplementary Figure 6. Raman spectra of PtSe$_2$ films made from different starting Pt film thickness showing the characteristic $E_g$ and $A_{1g}$ Raman-active modes for PtSe$_2$. The $A_{1g}/E_g$ intensity ratio increases with increasing layer thickness consistent with previous reports [S1].
Supplementary Figure 7. Typical two-point IV characteristic of a 5-6.5 nm PtSe₂ film indicating good Ohmic contacts. Linear behaviour was found for all samples.

Supplementary Table 1. Hall-effect results of two different batches of PtSe₂ samples grown separately with 3-month interval by e-beam evaporation of Pt film on Si/SiO₂, followed by TAC process at 400 °C, confirming the reproducibility of the process.

<table>
<thead>
<tr>
<th>Starting Pt film nominal thickness</th>
<th>1 nm Pt</th>
<th>1 nm Pt</th>
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<tbody>
<tr>
<td>Carrier Type</td>
<td>P</td>
<td>P</td>
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<tr>
<td>Hall mobility (cm²/V.s)</td>
<td>5.6</td>
<td>5.3</td>
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<tr>
<td>Sheet resistivity (Ω/sq)</td>
<td>2.1x10⁴</td>
<td>2.7x10⁴</td>
</tr>
<tr>
<td>Sheet carrier concentration (cm⁻²)</td>
<td>5.4x10¹³</td>
<td>4.3x10¹³</td>
</tr>
</tbody>
</table>

Supplementary Table 2. Hall-effect results of three batches of PtSe₂ samples grown by e-beam evaporation of Pt film on sapphire, followed by TAC process 400 °C showing very consistent results compared to Si/SiO₂ substrate.

<table>
<thead>
<tr>
<th>Starting Pt film nominal thickness</th>
<th>0.5 nm Pt</th>
<th>0.7 nm Pt</th>
<th>1 nm Pt</th>
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<tr>
<td>Carrier Type</td>
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<td>P</td>
<td>P</td>
<td>P</td>
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<tr>
<td>Hall mobility (cm²/V.s)</td>
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<td>5.0</td>
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<tr>
<td>Sheet resistivity (Ω/sq)</td>
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<td>5.6x10⁴</td>
<td>2.6x10⁴</td>
<td>6.1x10¹³</td>
</tr>
<tr>
<td>Sheet carrier concentration (cm⁻²)</td>
<td>1.5x10¹¹</td>
<td>2.7x10¹³</td>
<td>4.8x10¹¹</td>
<td>1.4x10¹⁴</td>
</tr>
</tbody>
</table>
(a) Drain Current, $I_{DS}$ (μA) vs. Gate Voltage, $V_{GS}$ (V)

- $I_{ON} / I_{OFF} = 230$
- $W_{ch} = 45$ μm
- $L_{ch} = 15$ μm
- $V_{DS} = 1$ V
- at room temp
- post transfer

2.5-3 nm PtSe$_2$

(b) Drain Current, $I_{DS}$ (μA) vs. Gate Voltage, $V_{GS}$ (V)

- $L_{ch} = 15$ μm -- $I_{ON} / I_{OFF} = 330$
- $L_{ch} = 35$ μm -- $I_{ON} / I_{OFF} = 175$
- $L_{ch} = 60$ μm -- $I_{ON} / I_{OFF} = 185$

- $W_{ch} = 20$ μm
- $V_{DS} = 1$ V
- at room temp
- post transfer

2.5-3 nm PtSe$_2$
Supplementary Figure 8. (a) Transfer characteristic of the same device as shown in Fig. 4(b), in semi-log scale.

Transfer characteristic of the back-gated FET devices with PtSe$_2$ channel thickness of (b) 2.5-3 nm, and (c) 5-6.5 nm, in semi-log scale.
Supplementary Figure 9. Activation energy ($E_A$) extracted from temperature-dependent measurements for (a) 2.5-3 nm PtSe$_2$ sample transferred onto unprocessed Si/SiO$_2$ substrate, (b) 2.5-3 nm PtSe$_2$ sample on as-grown substrate, and (c) 5-6.5 nm PtSe$_2$ sample on as-grown substrate.
Supplementary Figure 10. Variation of (left) $I_{\text{ON}}$ and $I_{\text{OFF}}$, and (right) $I_{\text{ON}}/I_{\text{OFF}}$ ratio with temperature for a typical device ($W_{\text{ch}} = 40 \, \mu\text{m}$ and $L_{\text{ch}} = 15 \, \mu\text{m}$) with 2.5-3 nm PtSe$_2$ channel thickness. $E_a$ is activation energy.

References