Supporting information

“Strain-dependent Magnetic Ordering Switching in 2D AFM Ternary V-based Chalcogenides Monolayers”

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Fig. S1 (a) The polytypic structures of VAl$_2$Se$_4$ monolayer for different H and T phases. (b) Relative energy ($\Delta E$) of different phases. Light red, yellow and light blue spheres represent V, Se and Al atoms, respectively.

Fig. S2. The phonon spectrums (a-c) and AIMD simulation (d-f) at 300K. (a) and (d) VAl$_2$Se$_4$ monolayer; (b) and (e) VAIGaSe$_4$ monolayers; (c) and (f) VGa$_2$Se$_4$ monolayer. The inset represents the configuration snapshots (one at the beginning and one at the end).

The Young’s modulus ($Y$) of 2D materials can be described in the form of polar coordinates, which can be expressed as\textsuperscript{1,2}

\[
Y(\theta) = \frac{c_{11}c_{22} - c_{12}^2}{c_{11} \sin^4 \theta + c_{22} - 2c_{12} \sin^2 \theta \cos^2 \theta + 2c_{12} \cos^2 \theta}
\]

where $c_{ij}$ and $\theta$ represent the elastic constants and the angle with respect to the x-axis, respectively.
Table S1. The mechanical properties of VXYSe₄(X, Y = Al, Ga). Calculated elastic constants (cᵢⱼ), Young’s modulus (Y in N/m).

<table>
<thead>
<tr>
<th>Structure</th>
<th>c₁₁</th>
<th>c₁₂</th>
<th>c₂₂</th>
<th>c₆₆</th>
<th>Y(0)/(N/m)</th>
</tr>
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<tbody>
<tr>
<td>VAl₂Se₄</td>
<td>69.683</td>
<td>22.027</td>
<td>69.683</td>
<td>0.135</td>
<td>62.720</td>
</tr>
<tr>
<td>VAlGaSe₄</td>
<td>66.930</td>
<td>22.633</td>
<td>66.930</td>
<td>0.239</td>
<td>59.276</td>
</tr>
<tr>
<td>VGa₂Se₄</td>
<td>65.748</td>
<td>21.083</td>
<td>65.748</td>
<td>0.272</td>
<td>58.987</td>
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</table>

Fig. S3 The planar average of the electrostatic potential of (a) VAl₂Se₄ monolayer; (b) VAlGaSe₄ monolayer; (c) VGa₂Se₄ monolayer. The purple, yellow, light red and light blue balls represent Ga, Se, V and Al atoms, respectively.

Fig. S4 AFM direct interactions of (a) d_yz-d_yz orbital exchange; (b) d_xz-d_xz orbital exchange; (c) d_yz-d_xy orbital exchange. (d) FM superexchange interactions of d-p-d orbital. The pink and green arrows represent spin-up and spin-down electron, respectively.

Fig. S5 The orbital-resolved band structure of (a) VAl₂Se₄ monolayer; (b) VAlGaSe₄ monolayers; (c) VGa₂Se₄ monolayer. The green, red, blue, orange and purple circles represent Se-p, Al-p, Ga-p, V-t₂g (dₓ²−y², dₓz, dᵧz) and V-e₉ (dₓ²−y², dz²), respectively. The spin-up and spin-down channels are expressed by the pink and cyan arrows.
Fig. S6 The electronic band structure of 2D VXYSe$_4$ (X, Y = Al, Ga) with spin-orbit coupling (SOC). (a) VAl$_2$Se$_4$; (b)VAlGaSe$_4$; (c)VGa$_2$Se$_4$.

Table S2 The band gap of 2D VXYSe$_4$ (X, Y = Al, Ga) with and without spin-orbit coupling (SOC)

<table>
<thead>
<tr>
<th></th>
<th>VAl$_2$Se$_4$</th>
<th>VAlGaSe$_4$</th>
<th>VGa$_2$Se$_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>PBE+U (eV)</td>
<td>0.8625</td>
<td>0.4276</td>
<td>0.3266</td>
</tr>
<tr>
<td>PBE+U+SOC (eV)</td>
<td>0.8614</td>
<td>0.4271</td>
<td>0.3231</td>
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</table>

**Part I. Calculations of the magnetic interaction parameters $J$ and $A$.**

According to the Heisenberg model, the four different magnetic configurations in Figure.1(c-f) can be expressed as

\[
\begin{align*}
E_{F\text{M}} &= E_0 + 4S^2(-6J_1 - 6J_2) \\
E_{AF\text{M}1} &= E_0 + 4S^2(2J_1 + 2J_2) \\
E_{AF\text{M}2} &= E_0 + 4S^2(2J_1 - 2J_2) \\
E_{AF\text{M}3} &= E_0 + 4S^2(-2J_1 + 2J_2)
\end{align*}
\]

(1.1)

Here $E_0$ is total energy of the system without magnetic interactions, $|S| = \frac{3}{2}$ for V cation.

Fig. S7. The (a) bond length and (b) bond angle of 2D VXYSe$_4$ (X, Y = Al, Ga) monolayers as a function of strain.
Fig. S8 The band structure of VAl$_2$Se$_4$ monolayer under biaxial strains of (a) -6%, (b) -4%, (c) -2%, (d) 2%, (e) 4%, (f) 6%. The spin-up and spin-down channels are expressed by the blue and red lines, respectively.

Fig. S9 The band structure of VAlGaSe$_4$ monolayer under biaxial strains of (a) -6%, (b) -4%, (c) -2%, (d) 2%, (e) 4%, (f) 6%. The spin-up and spin-down channels are expressed by the blue and red lines, respectively.
Fig. S10 The band structure of VGa$_2$Se$_4$ monolayer under biaxial strains of (a) -6%, (b) -4%, (c) -2%, (d) 2%, (e) 4%, (f) 6%. The spin-up and spin-down channels are expressed by the blue and red lines, respectively.

Fig. S11. The first nearest-neighboring magnetic exchange parameters (purple line) and the second nearest-neighboring magnetic exchange parameters (orange line) of 2D (a) VAl$_2$Se$_4$; (b) VAlGaSe$_4$; (c) VGa$_2$Se$_4$ monolayers under different strain.

**Reference**