Supporting information

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

Kaijuan Pang[a], Xiaodong Xu[b*], Yadong Wei[b], Tao Ying[a], Bo Gao[a], Weiqi Li[a*], Yongyuan Jiang[a, c, d *]

[a] School of Physics, Harbin Institute of Technology, Harbin 150001, China

[b] School of Material Science and Engineering, Harbin Institute of Technology, Harbin 150001, China

[c] Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan 030006, China

[d] Key Lab of Micro-Optics and Photonic Technology of Heilongjiang Province, Harbin 150001, China

Email: xuxd@hit.edu.cn

Email: jiangyy@hit.edu.cn

Email: tccliweiqi@hit.edu.cn

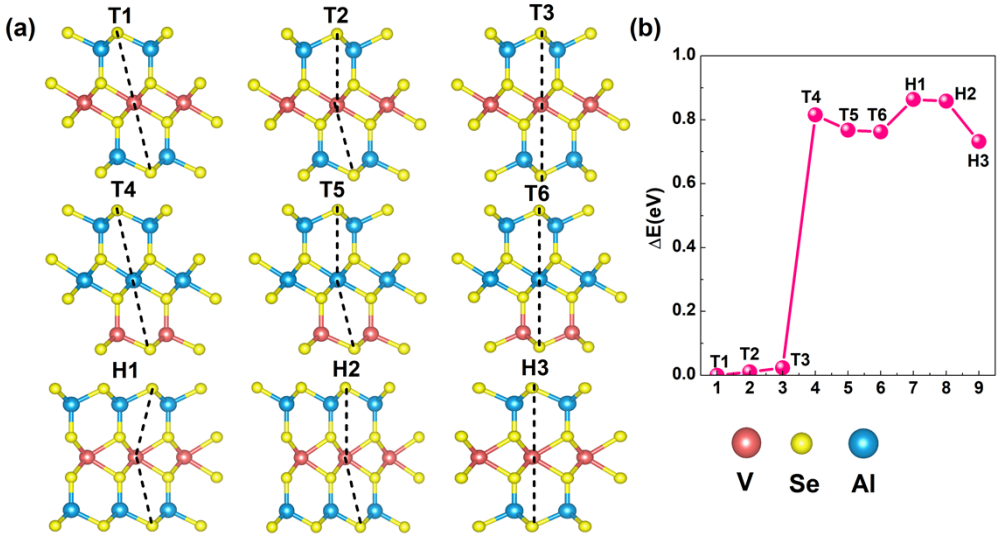

Fig.S1 (a) The polytypic structures of VAl_2Se_4 monolayer for different H and T phases. (b) Relative energy (ΔE) of different phases. Light red, yellow and light blue spheres represent V, Se and Al atoms, respectively.

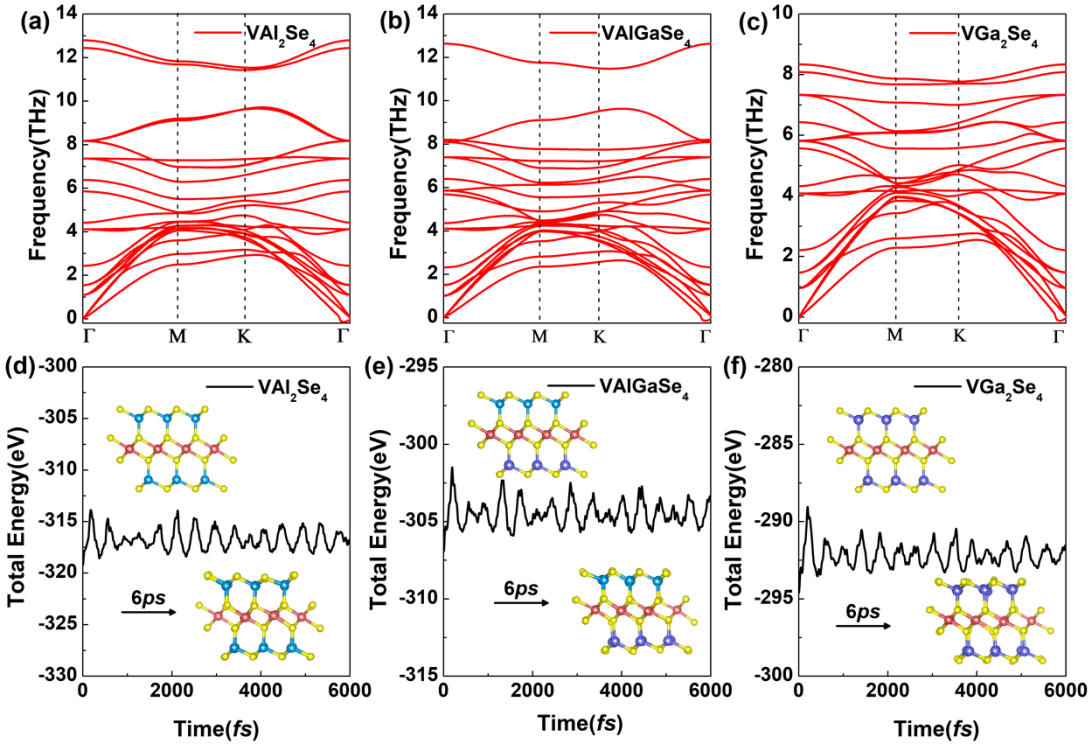

Fig.S2. The phonon spectrums (a-c) and AIMD simulation (d-f) at 300K. (a) and (d) VAl_2Se_4 monolayer; (b) and (e) $VAlGaSe_4$ monolayers; (c) and (f) VGa_2Se_4 monolayer. The inset represents the configuration snapshots (one at the beginning and one at the end).

The Young's modulus (Y) of 2D materials can be described in the form of polar coordinates, which can be expressed as[1, 2]

$$Y(\theta) = \frac{c_{11}c_{22} - c_{12}^2}{c_{11}\sin^4\theta[c_{11}c_{22} - c_{12}^2)/c_{66} - 2c_{12}]\sin^2\theta\cos^2\theta + c_{22}\cos^4\theta}$$

where c_{ij} and θ represent the elastic constants and the angle with respect to the ***x***-axis, respectively.

Table.S1. The mechanical properties of $VXYSe_4$ (X, Y =Al, Ga). Calculated elastic constants (c_{ij}), Young's modulus (Y in N/m).

Structure	c_{11}	c_{12}	c_{22}	c_{66}	Y(0)/(N/m)
VAl_2Se_4	69.683	22.027	69.683	0.135	62.720
$VAlGaSe_4$	66.930	22.633	66.930	0.239	59.276
VGa_2Se_4	65.748	21.083	65.748	0.272	58.987

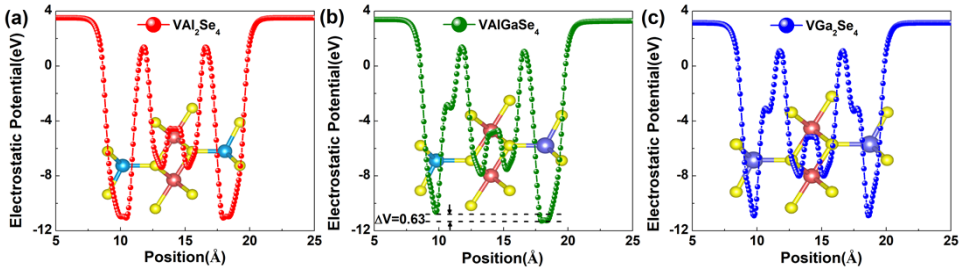

Fig.S3 The planar average of the electrostatic potential of (a) VAl_2Se_4 monolayer; (b) $VAlGaSe_4$ monolayer; (c) VGa_2Se_4 monolayer. The purple, yellow, light red and light blue balls represent Ga, Se, V and Al atoms, respectively.

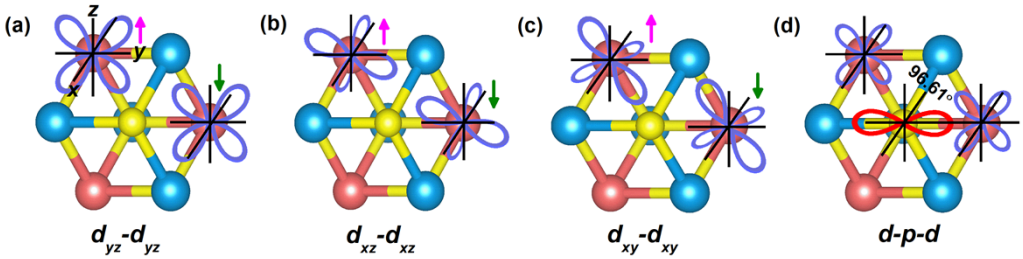

Fig.S4 AFM direct interactions of (a) d_{yz}-d_{yz} orbital exchange; (b) d_{xz}-d_{xz} orbital exchange; (c) d_{xy}-d_{xy} orbital exchange. (d) FM superexchange interactions of *d*-*p*-*d* orbital. The pink and green arrows represent spin-up and spin-down electron, respectively.

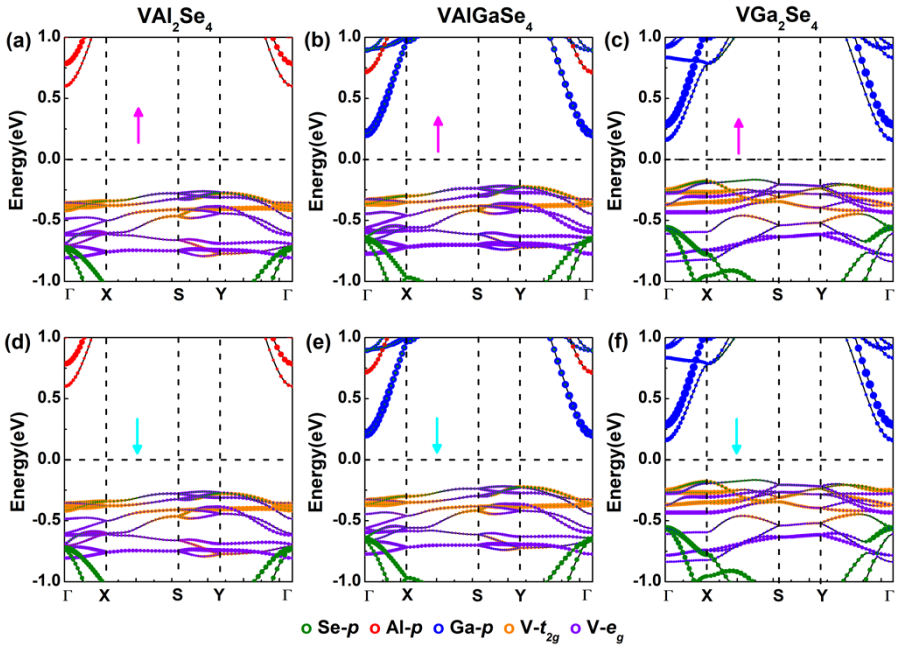

Fig.S5 The orbital-resolved band structure of (a) VAl_2Se_4 monolayer; (b) $VAlGaSe_4$ monolayers; (c) VGa_2Se_4 monolayer. The green, red, blue, orange and purple circles represent Se-*p*, Al-*p*, Ga-*p*, V-t_{2g} (d_{xy}, d_{yz}, d_{xz}) and V-e_g (dx^2-y^2, dz^2), respectively. The spin-up and spin-down channels are expressed by the pink and cyan arrows.

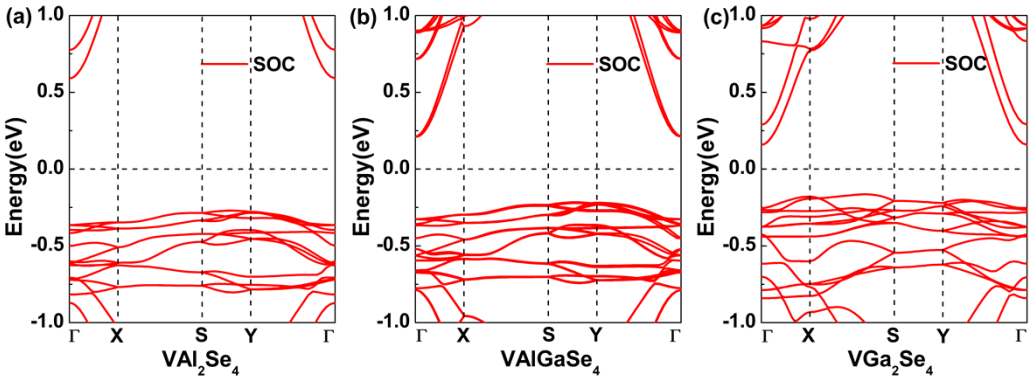

Fig.S6 The electronic band structure of 2D $VXYSe_4$ (X, Y = Al, Ga) with spin-orbit coupling (SOC). (a) VAl_2Se_4; (b)$VAlGaSe_4$; (c)VGa_2Se_4.

Table.S2 The band gap of 2D $VXYSe_4$ (X, Y = Al, Ga) with and without spin-orbit coupling (SOC)

	VAl_2Se_4	$VAlGaSe_4$	VGa_2Se_4
PBE+U (eV)	0.8625	0.4276	0.3266
PBE+U+SOC (eV)	0.8614	0.4271	0.3231

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

$$\begin{aligned} E_{FM} &= E_0 + 4S^2(-6J_1 - 6J_2) \\ E_{AFM1} &= E_0 + 4S^2(2J_1 + 2J_2) \\ E_{AFM2} &= E_0 + 4S^2(2J_1 - 2J_2) \\ E_{AFM3} &= E_0 + 4S^2(-2J_1 + 2J_2) \\ A &= Y - MAE = E_{[001]} - E_{[010]} \\ X - MAE &= E_{[001]} - E_{[100]} \end{aligned} \tag{1.1}$$

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

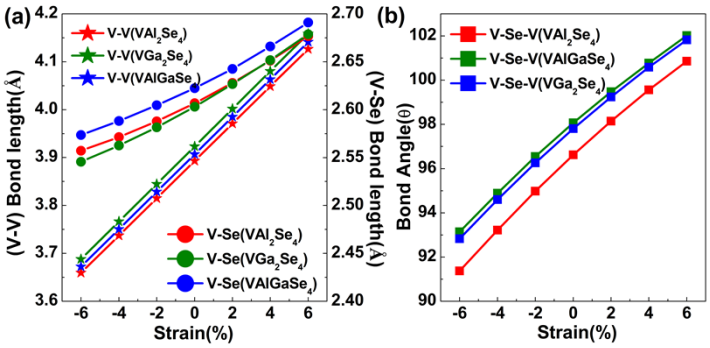

Fig.S7. The (a) bond length and (b) bond angle of 2D $VXYSe_4$ (X, Y = Al, Ga) monolayers as a function of strain.

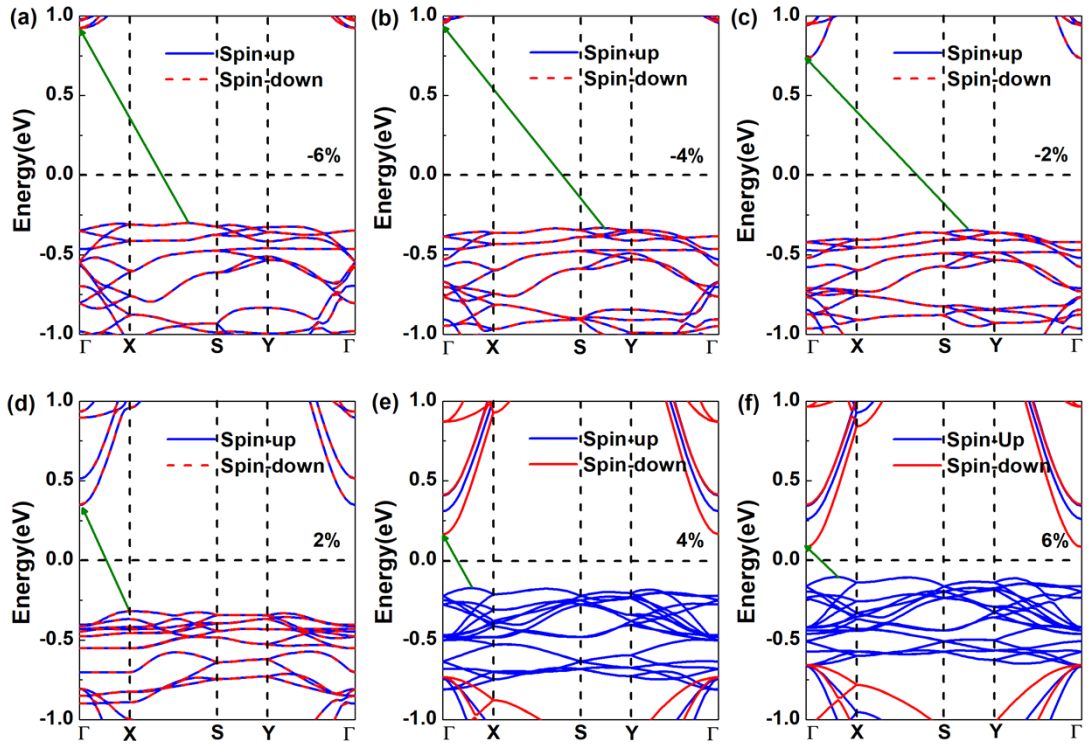

Fig.S8 The band structure of VAl_2Se_4 monolayer under biaxial strains of (a) -6%, (b) -4%, (c) -2%, (d) 2%, (e) 4%, (f) 6%. The spin-up and spin-down channels are expressed by the blue and red lines, respectively.

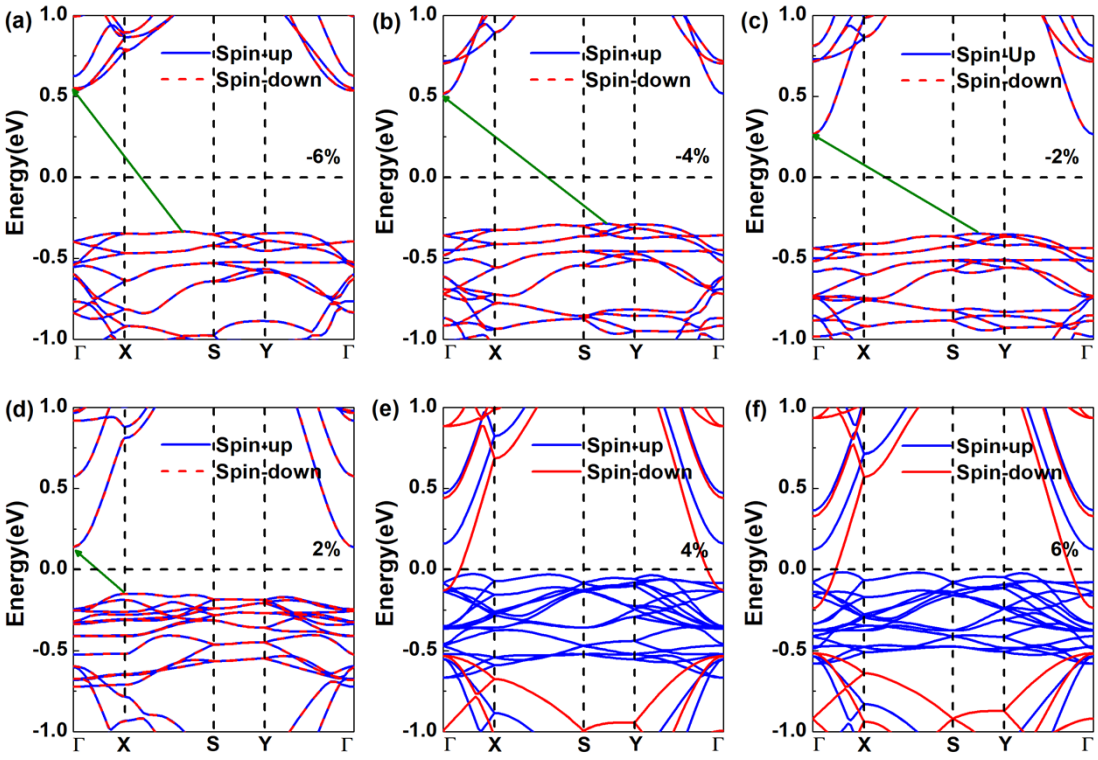

Fig.S9 The band structure of $VAlGaSe_4$ monolayer under biaxial strains of (a) -6%, (b) -4%, (c) -2%, (d) 2%, (e) 4%, (f) 6%. The spin-up and spin-down channels are expressed by the blue and red lines, respectively.

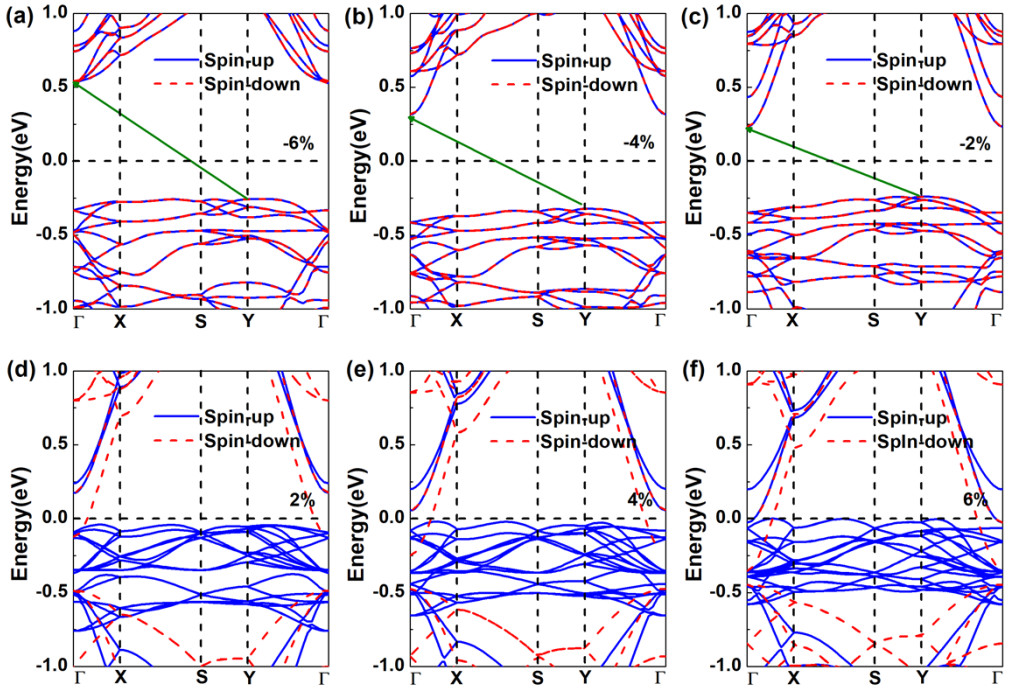

Fig.S10 The band structure of VGa_2Se_4 monolayer under biaxial strains of (a) -6%, (b) -4%, (c) -2%, (d) 2%, (e) 4%, (f) 6%. The spin-up and spin-down channels are expressed by the blue and red lines, respectively.

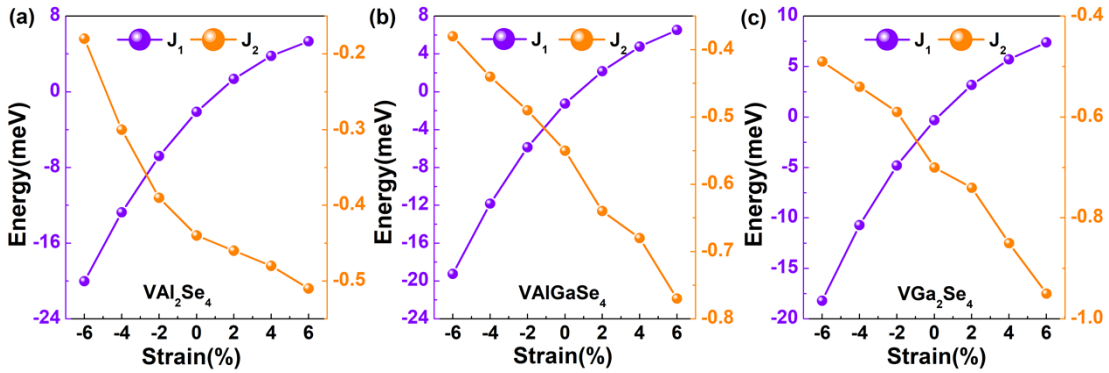

Fig.S11. The first nearest-neighboring magnetic exchange parameters (purple line) and the second nearest-neighboring magnetic exchange parameters (orange line) of 2D (a) VAl_2Se_4; (b) $VAlGaSe_4$; (c) VGa_2Se_4 monolayers under different strain.

Reference

1. E. Bolen and E. Deligoz, *J. Appl. Phys*, 2021, **130**, 065102
2. M. Sun and U. Schwingenschlögl, *Adv. Energy Mater*, 2021, **11**, 2003633.