Supporting Information for 'Tensile strain induced switching of magnetic states in $NbSe_2$ and NbS_2 single layers'

Ying Xu, Xiaofei Liu and Wanlin Guo*

State Key Laboratory of Mechanics and Control of Mechanical Structures, Key Laboratory for Intelligent Nano Materials and Devices of Ministry of Education, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China

*Address correspondence to wlguo@nuaa.edu.cn.

This file includes: Additional Figures S1-S4, TABLE S1-S2 and Estimation of critical temperatures.

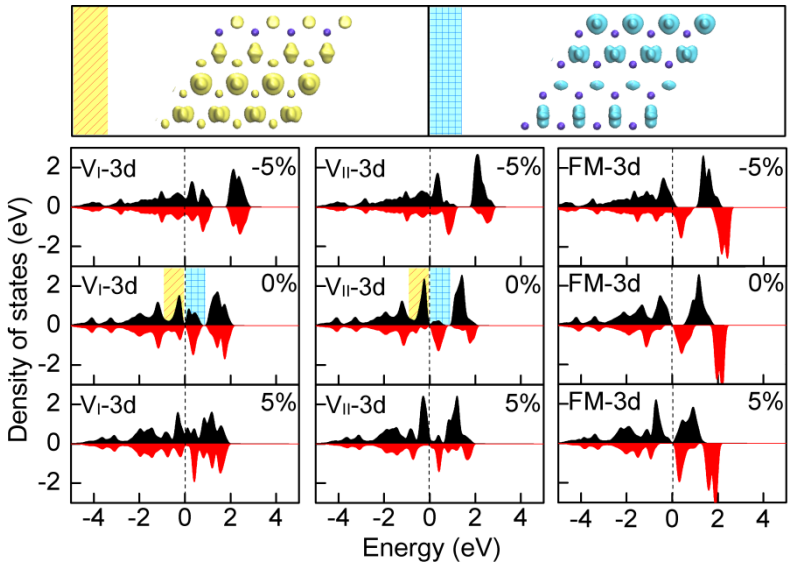

Figure S1. Spin-polarized density of states of VSe_2 under -5%, 0% and 5% biaxial tensile strains. The black (red) regions represent the spin-up (spin-down) components. V_I and V_{II} are the V atoms of low and high magnetic moment in the AFM spin configuration. The Fermi level is set to zero. The spin-up states of VSe_2 under 3% strain shown by the isosurface plots (0.05 eV/Å^3) are within the energy range of -1~0 eV and 0~1 eV respectively.

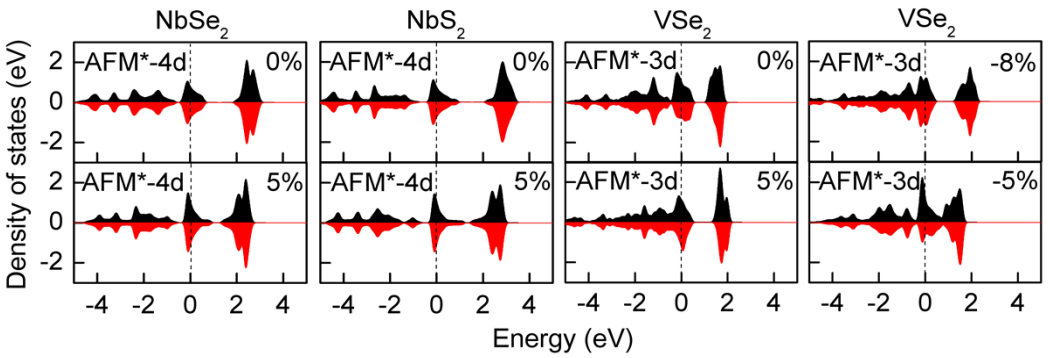

Figure S2. Spin-polarized density of AFM* states (DOS) in $NbSe_2$, NbS_2 and VSe_2 under 0% and 5% biaxial tensile strain and VSe_2 under -8% and -5% strain. The black (red) regions represent the spin-up (spin-down) components. The Fermi level is set to zero.

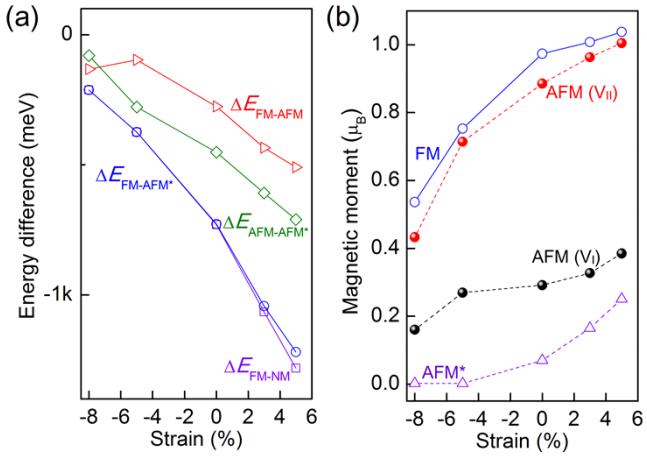

Figure S3. Energy differences per unit cell (a) and the magnetic moment per V atom (b) in different magnetic state of singled-layered VS_2. The solid lines in (b) denote the magnetic moment of the magnetic ground state and the dot lines represent that in the less stable magnetic state. V_I and V_{II} denotes the V atoms with the lower and high magnetic moment in the AFM state.

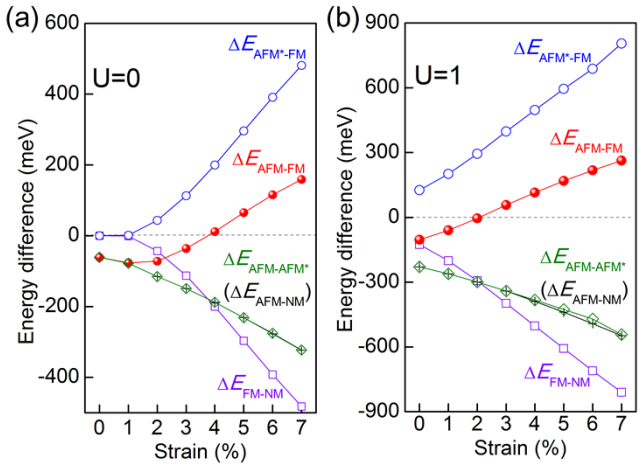

Figure S4. Energy differences between different magnetic states in singled-layered $NbSe_2$ as a fucntion of strain for (a) U=0 eV and (b) U=1 eV.

TABLE S1. The optimized structure parameters (lattice constant, a; M-X bond length, dM-X; X-X bond length dX-X) in MX2 (M = Nb, V; X=Se, S) single layers.

	a (Å)	$d_{M\text{-}X}$ (Å)	$d_{X\text{-}X}$ (Å)
$NbSe_2$	3.486	2.626	3.354
NbS_2	3.350	2.491	3.140
VSe_2	3.331	2.495	3.178
VS_2	3.174	2.355	2.957

Estimation of critical temperatures

According to Heisenberg model, the Hamiltonian can be written as:

$$H = -\sum_{i,j,i\neq j} J\vec{S}_i \cdot \vec{S}_j$$

Where, S_i is the net spin in the ith cation, J is the exchange coupling constant.

In this work, we will consider the nearest and next nearest interactions.

In $NbSe_2$ system, there are six nearest neighbors and six next nearest neighbors both for ferro- and antiferro-magnetic states.

$$E_{FM} = -6NJ_{F1}S^2 - 6NJ_{F2}S^2$$

$$E_{AFM} = -N\left(J_{A1}S_1^2 + J_{A2}S_2^2\right) - N\left(-J_{A3}S_1^2 - J_{A4}S_2^2\right)$$
$$= -N\left(J_{A1}S_1^2 + J_{A2}S_2^2 - J_{A3}S_1^2 - J_{A4}S_2^2\right)$$

Where J_{F1}, J_{A1} and J_{A2} are the nearest exchange coupling constant between different cations, while J_{F2}, J_{A3} and J_{A4} are the next nearest exchange coupling constant between different cations. Therefore, the energy difference between AFM and FM state is,

$$\begin{aligned}\Delta E &= E_{AFM} - E_{FM} \\ &= -N\left(J_{A1}S_1^2 + J_{A2}S_2^2 - 6J_{F1}S^2\right) + N\left(J_{A3}S_1^2 + J_{A4}S_2^2 + 6J_{F2}S^2\right)\end{aligned}$$

In the equation of the energy difference, the first item is the nearest intercation, while the second item is the next nearest interaction. Obviously, it is difficult to estimate the Curie temperature with so many different exchange energy J_i (i = F1, F2 and A1 to A4). To obtain a rough estimation of Curie temperature, we neglect the next nearest interaction and suppose that the exchange energy and spin magnetic moment are almost the same for ferro- and antiferro-magnetic states,

$$J_{A1} = J_{A2} = J_{FM} = J, \left|S_1\right| = \left|S_2\right| = S$$

Therefore, we can obtain the relation between exchange energy and the energy difference,

$$J = \Delta E \Big/ 4NS^2$$

Where N is the total number of magnetic atoms,

$$S = \mu_S \Big/ 2\mu_B$$

Where μ_S is the magnetic moment per atom, μ_B is the Bohr magneton.

In mean field theory, the Curie temperature can be estimated by,

$$T = \frac{2}{3}\frac{J}{k_B}$$

Where k_B is the Boltzmann constant.

TABLE S2. The estimated Curie temperature under free strain and 7% biaxial tensile strain in $NbSe_2$ single layer.

	ΔE (meV)	J (meV)	T_C (K)
0%	-61.12	-61.86	-478.14
7%	159.02	25.12	194.16