Supporting Information for ‘Tensile strain induced switching of magnetic states in NbSe₂ and NbS₂ single layers’

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

Figure S1. Spin-polarized density of states of VSe₂ under -5%, 0% and 5% biaxial tensile strains. The black (red) regions represent the spin-up (spin-down) components. V₁ and V₁I 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 VSe₂ under 3% strain shown by the isosurface plots (0.05 eV/Å³) 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 single-layered NbSe$_2$ as a function 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, $d_{M-X}$; X-X bond length $d_{X-X}$) in MX$_2$ (M = Nb, V; X=Se, S) single layers.

<table>
<thead>
<tr>
<th></th>
<th>$a$ (Å)</th>
<th>$d_{M-X}$ (Å)</th>
<th>$d_{X-X}$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NbSe$_2$</td>
<td>3.486</td>
<td>2.626</td>
<td>3.354</td>
</tr>
<tr>
<td>NbS$_2$</td>
<td>3.350</td>
<td>2.491</td>
<td>3.140</td>
</tr>
<tr>
<td>VSe$_2$</td>
<td>3.331</td>
<td>2.495</td>
<td>3.178</td>
</tr>
<tr>
<td>VS$_2$</td>
<td>3.174</td>
<td>2.355</td>
<td>2.957</td>
</tr>
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</table>

Estimation of critical temperatures

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

$$ H = - \sum_{i,j,i \neq j} J S_i \cdot S_j $$

Where, $S_i$ is the net spin in the $i$th 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,

$$\Delta E = E_{AFM} - E_{FM}$$

$$= -N \left( J_{A1}S_1^2 + J_{A2}S_2^2 - 6J_{F1}S_1^2 \right) + N \left( J_{A3}S_1^2 + J_{A4}S_2^2 + 6J_{F2}S_1^2 \right)$$

In the equation of the energy difference, the first item is the nearest interaction, 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 antiferromagnetic states,

$$J_{A1} = J_{A2} = J_{FM} = J_s |S_1| = |S_2| = S$$

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

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

Where $N$ is the total number of magnetic atoms,

$$S = \frac{\mu_S}{2\mu_B}$$

Where $\mu_S$ is the magnetic moment per atom, $\mu_B$ is the Bohr magneton.

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

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

Where $k_B$ is the Boltzmann constant.

<table>
<thead>
<tr>
<th>Table S2. The estimated Curie temperature under free strain and 7% biaxial tensile strain in NbSe$_2$ single layer.</th>
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<tbody>
<tr>
<td>$\Delta E$ (meV)</td>
</tr>
<tr>
<td>------------------</td>
</tr>
<tr>
<td>0%</td>
</tr>
<tr>
<td>7%</td>
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