Fig. S1 shows angle dependencies of the total energy and magnetic moment in the case of the rotation of the whole Se upper layer of VSe₂ monolayer (a,b) and bulk (c,d) within the plane scheme.

![Graphs](image)

**Fig. S1** Total energy (a,c) and magnetic moment (b,d) as functions of the rotation angles obtained within the plane model for all Se atoms belonging to the upper layer. The simulations were performed for VSe₂ monolayer (a,b) and bulk (c,d).

For monolayer one can see that such a rotation scheme is less profitable in energy, since the barrier grows. In turn, the magnetic moment demonstrates the same behavior as with the arc rotation model. In the bulk case we obtain almost the same dependencies, but the maximum of the energy barrier at 30° becomes larger than that in the case of the monolayer.

Also we calculated partial densities of states of VSe₂ bulk in intermediate points of arc type of rotation with the 20° step (Fig S2).

**Fig. S2** Partial densities of states calculated for VSe₂ bulk in the ferromagnetic configuration. The arc rotation scheme with the 20° step was used. Left and right panels correspond to \((d_{xy}, d_{yz}, d_{xz})\) and \((d_{x^2-y^2}, d_{z^2})\) sets of states, respectively.

Figures S3 and Fig. S4 give band structures of VSe₂ monolayer and bulk obtained within arc scheme rotation with the 10° elementary step.
Fig. S3 Band structures of the monolayer VSe$_2$ calculated for atomic structures modified within the arc rotation model from T phase (0°) to H phase (60°) with the step of 10°. All the calculations were performed for ferromagnetic configuration. Red lines correspond to spin up states and black ones to spin down.

Fig. S4 Band structures of the bulk VSe$_2$ crystal calculated for atomic structures modified within the arc rotation model from T phase (0°) to H phase (60°) with the step of 10°. All the calculations were performed for ferromagnetic configuration. Red lines correspond to spin up states and black ones to spin down.