

## Supporting information

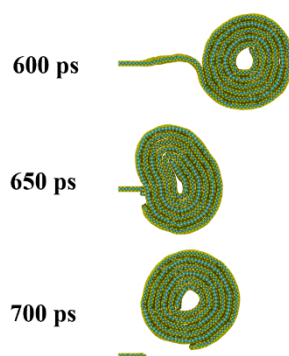
# Spontaneous Formation of MoS<sub>2</sub> Nanoscroll from Flat Monolayer with Sulfur Vacancies: A Molecular Dynamics Investigation

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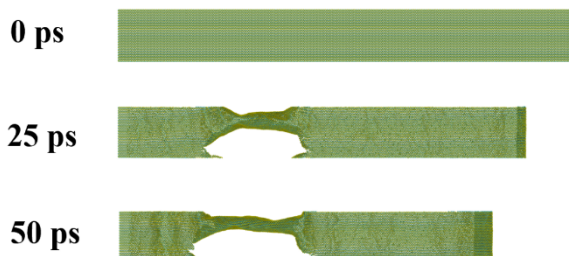
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Armchair nanoribbon with 20% vacancy density

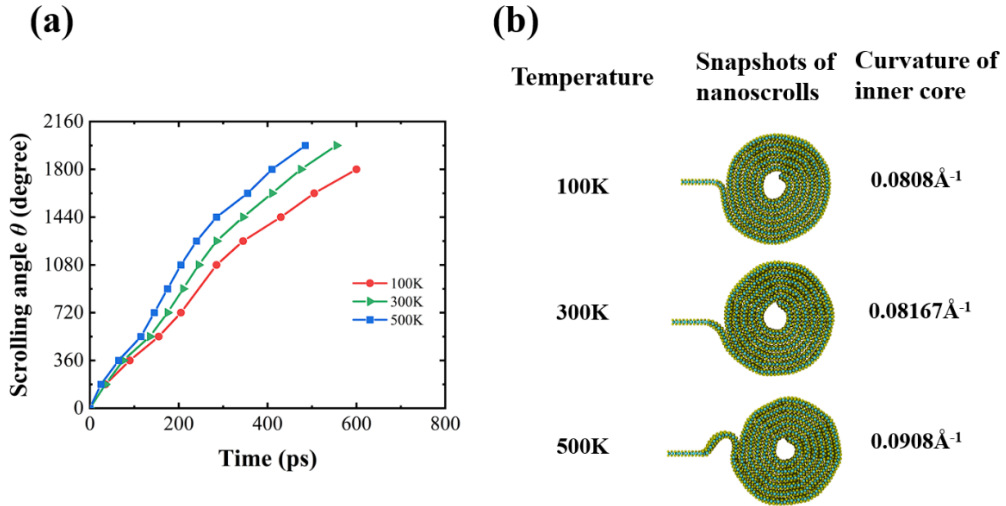


**Fig. S1** MD snapshots of the armchair nanoribbon with 20% vacancy density at 600 ps, 650 ps, and 700 ps.

Armchair nanoribbon with 35% vacancy density

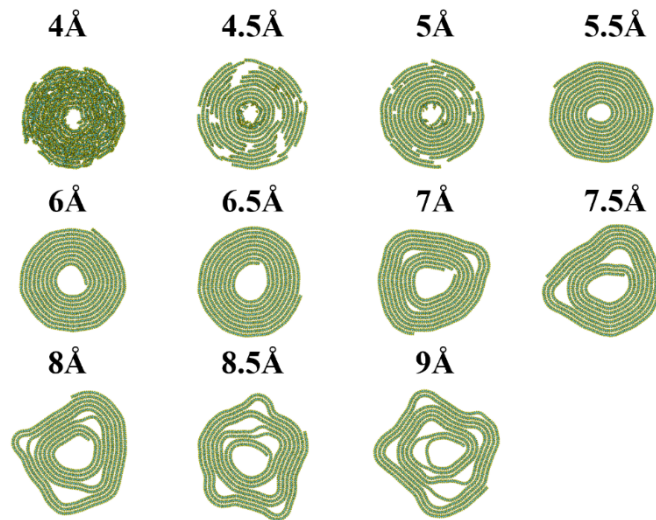


**Fig. S2** MD snapshots of the armchair nanoribbon with 35% vacancy density at 0 ps, 25 ps and 50 ps.

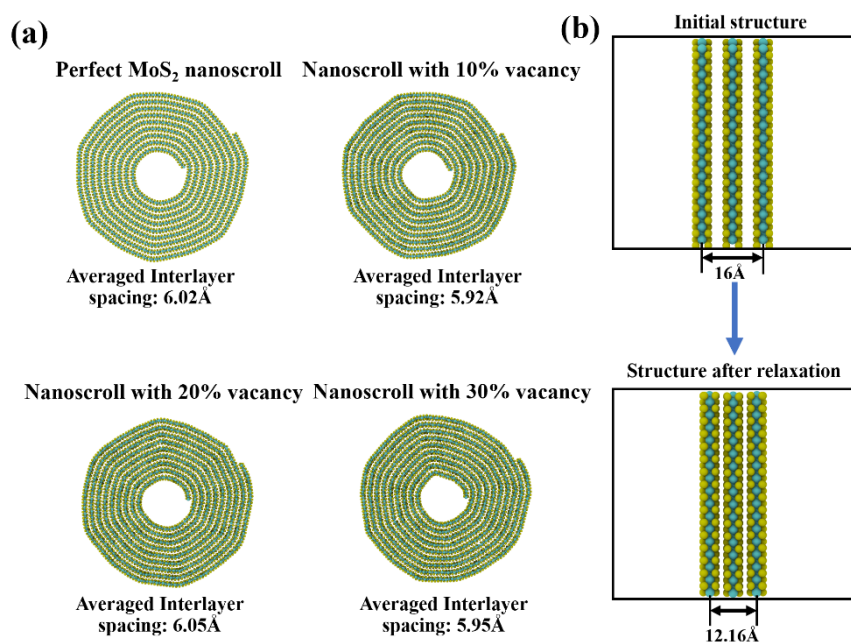


**Fig. S3** (a) The scrolling angle  $\theta$  with simulation time for zigzag nanoribbon with 15% sulfur vacancy at various simulation temperatures. (b) Structural snapshots of nanoscroll formation at various temperatures.

**Perfect MoS<sub>2</sub> nanoscrolls with different layer spacings**



**Fig. S4** MD simulation of perfect MoS<sub>2</sub> nanoscrolls with varying layer spacing, ranging from 4 Å to 9 Å.



**Fig. S5** (a) MoS<sub>2</sub> nanoscrolls with varying sulfur vacancy densities. (b) Planar multilayer MoS<sub>2</sub>, before and after relaxation of interlayer spacing.