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Supporting Information

Stability, structure and reconstruction of 1H-edges in MoS₂

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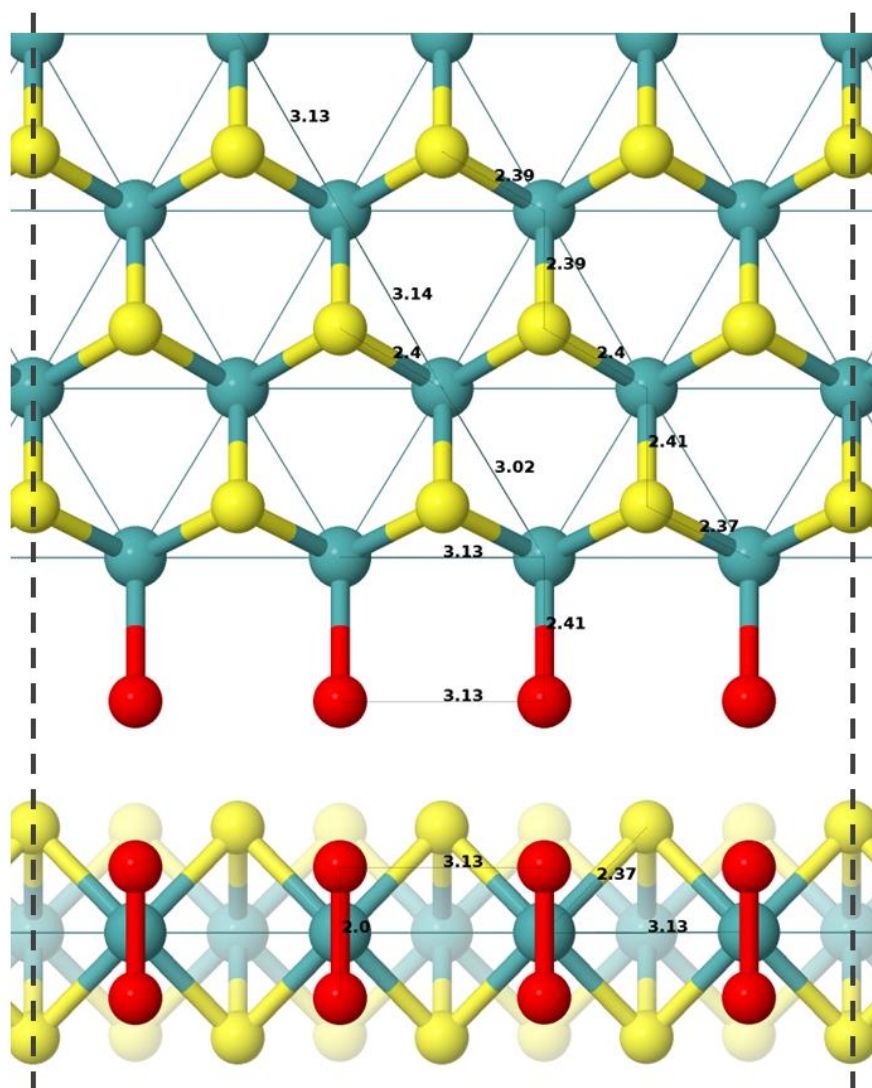
Mo-100%S configurations (Figures S1-S6)

Figure S1 Top and side views of Mo-100%S (VD). Grey dashed lines indicate the length of the simulation cell. Atomic distances are in Angstroms. Mo-Mo and Mo-S bulk values are 3.13 Å and 2.39 Å respectively. Atom colour code: blue-green (Mo), yellow (S), red (edge S).

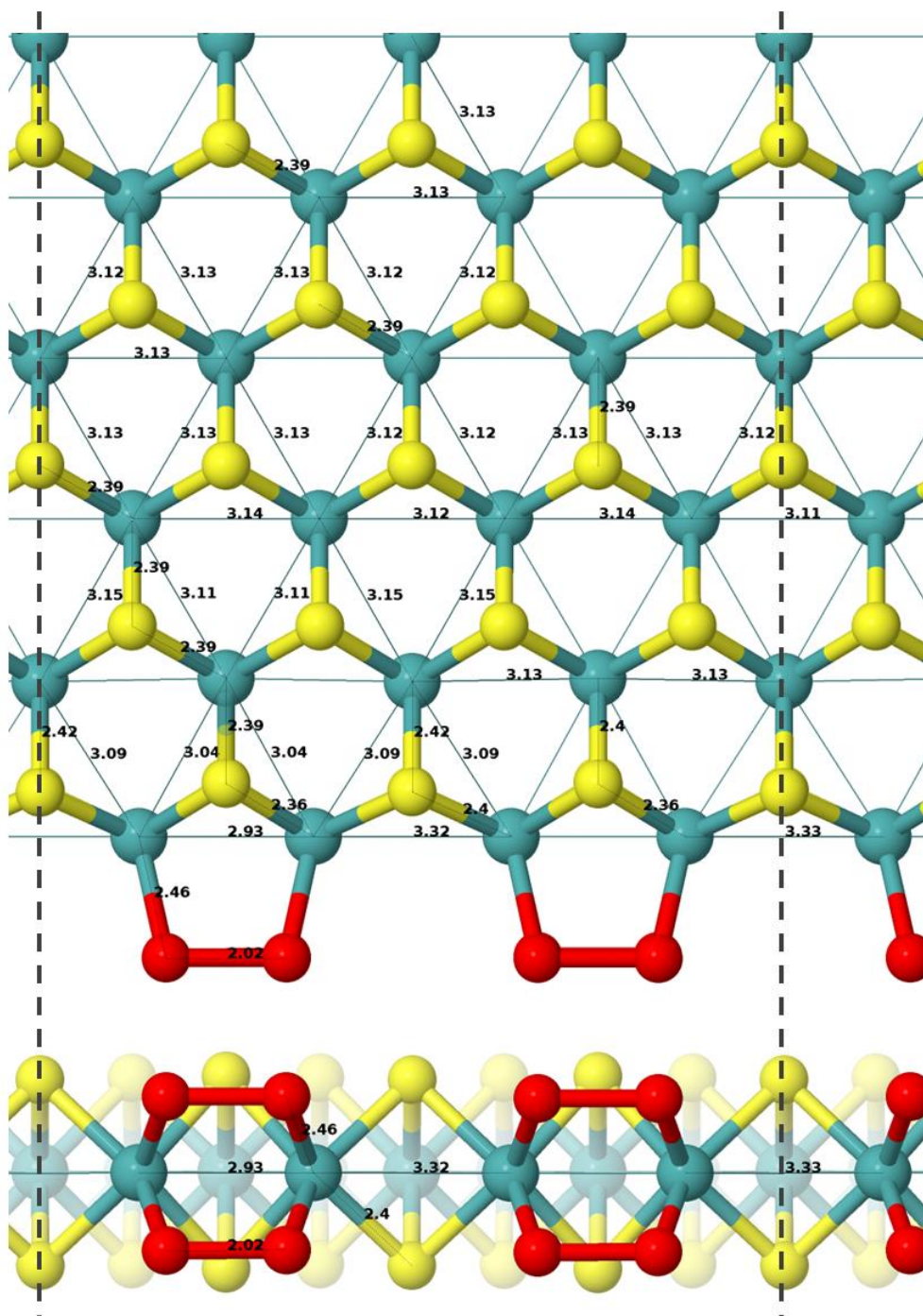


Figure S2 Top and side views of Mo-100%S (HD-O). Grey dashed lines indicate the length of the simulation cell. Atomic distances are in Angstroms. Mo-Mo and Mo-S bulk values are 3.13 Å and 2.39 Å respectively. Atom colour code: blue-green (Mo), yellow (S), red (edge S).

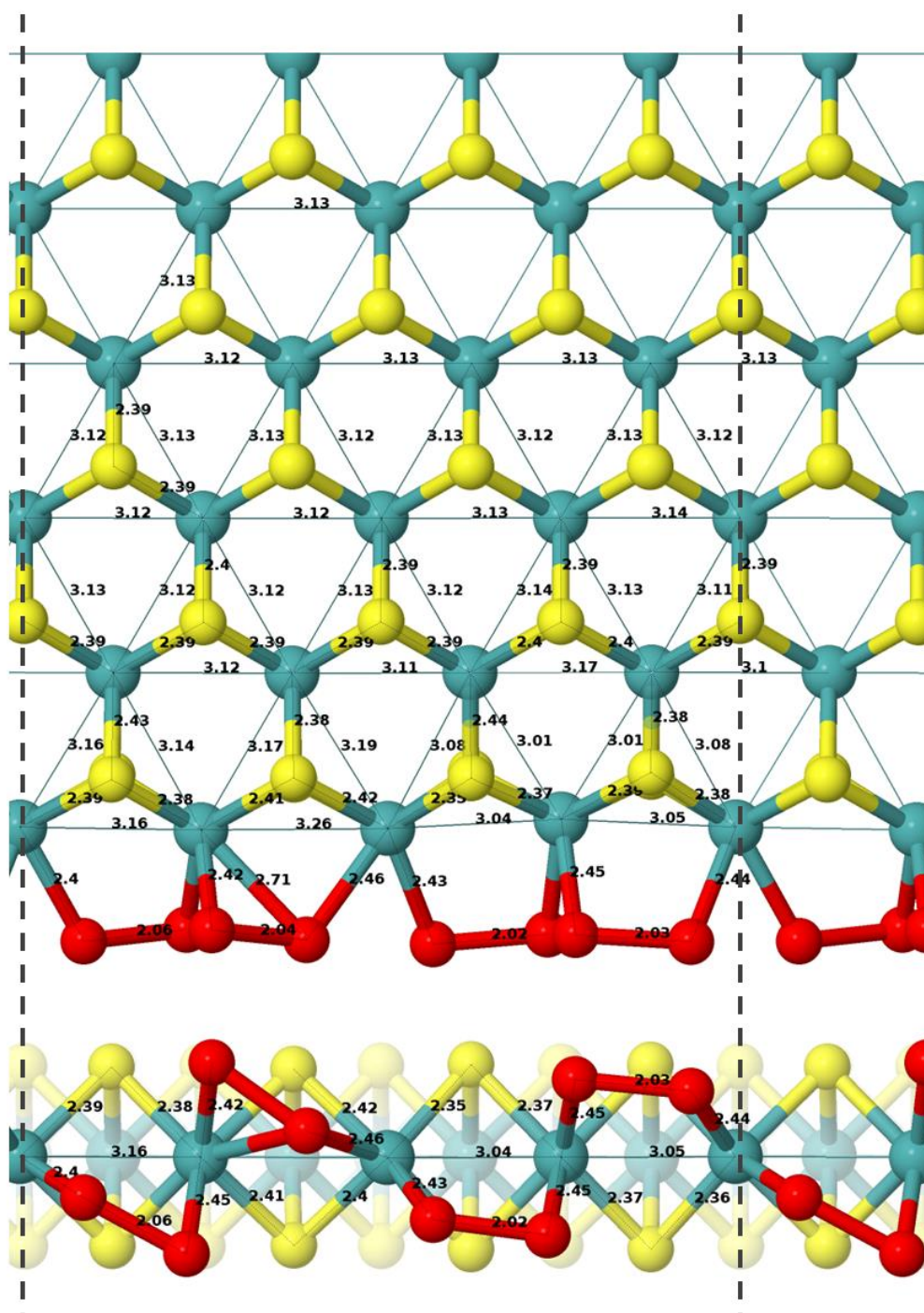
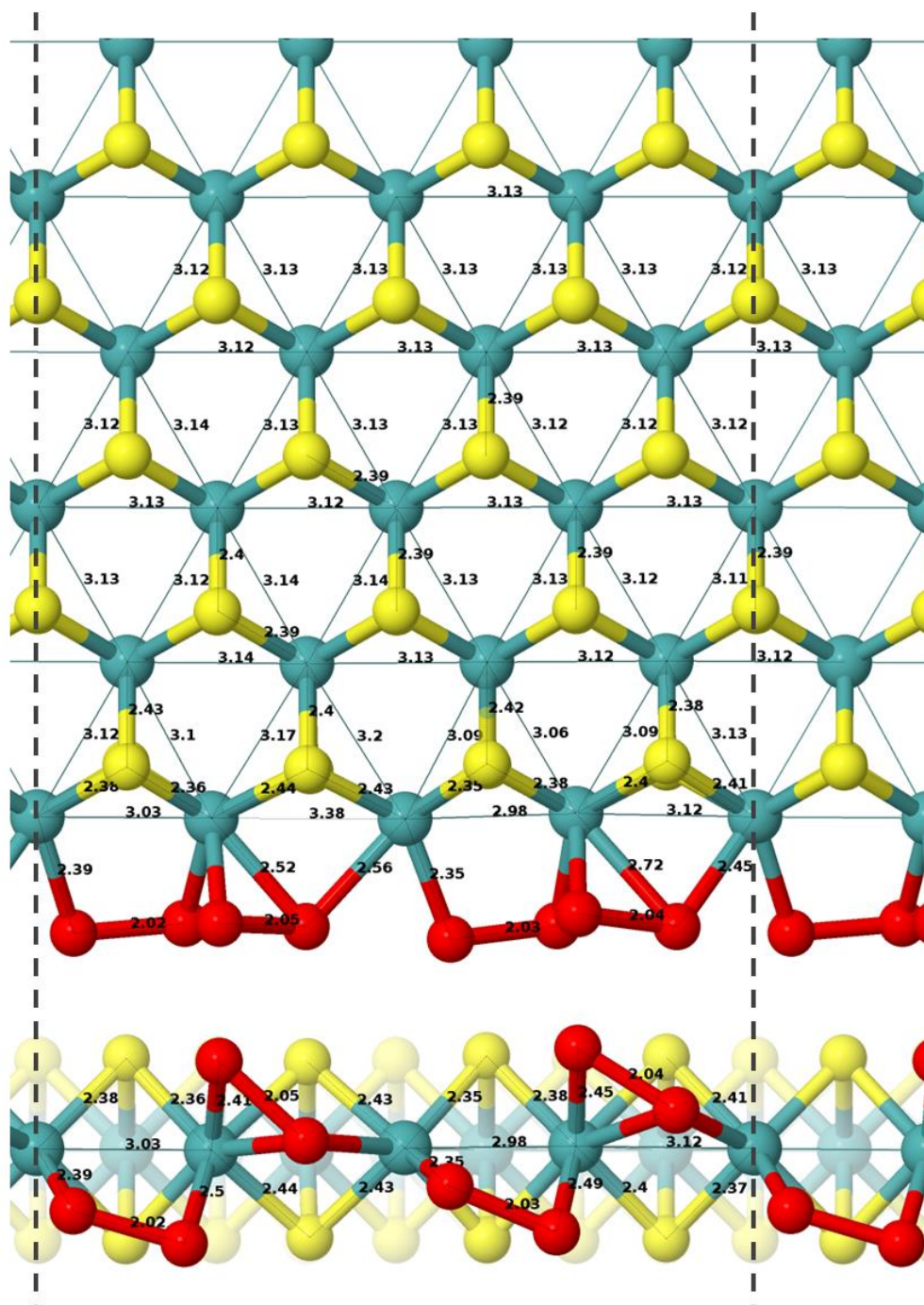


Figure S3 Top and side views of Mo-100%S (HD-S). Grey dashed lines indicate the length of the simulation cell. Atomic distances are in Angstroms. Mo-Mo and Mo-S bulk values are 3.13 Å and 2.39 Å respectively. Atom colour code: blue-green (Mo), yellow (S), red (edge S).



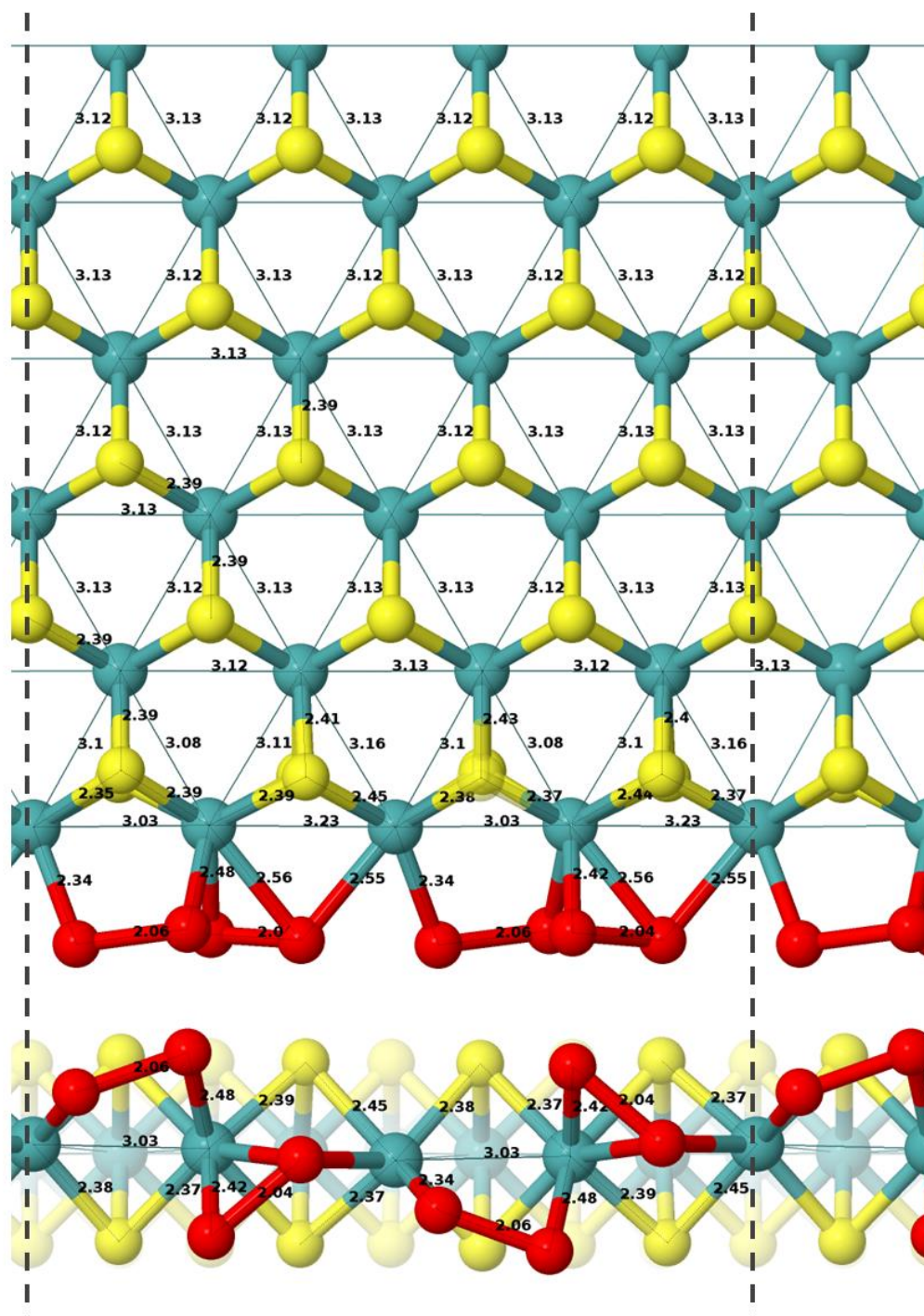


Figure S5 Top and side views of Mo-100%S (R2). Grey dashed lines indicate the length of the simulation cell. Atomic distances are in Angstroms. Mo-Mo and Mo-S bulk values are 3.13 Å and 2.39 Å respectively. Atom colour code: blue-green (Mo), yellow (S), red (edge S).

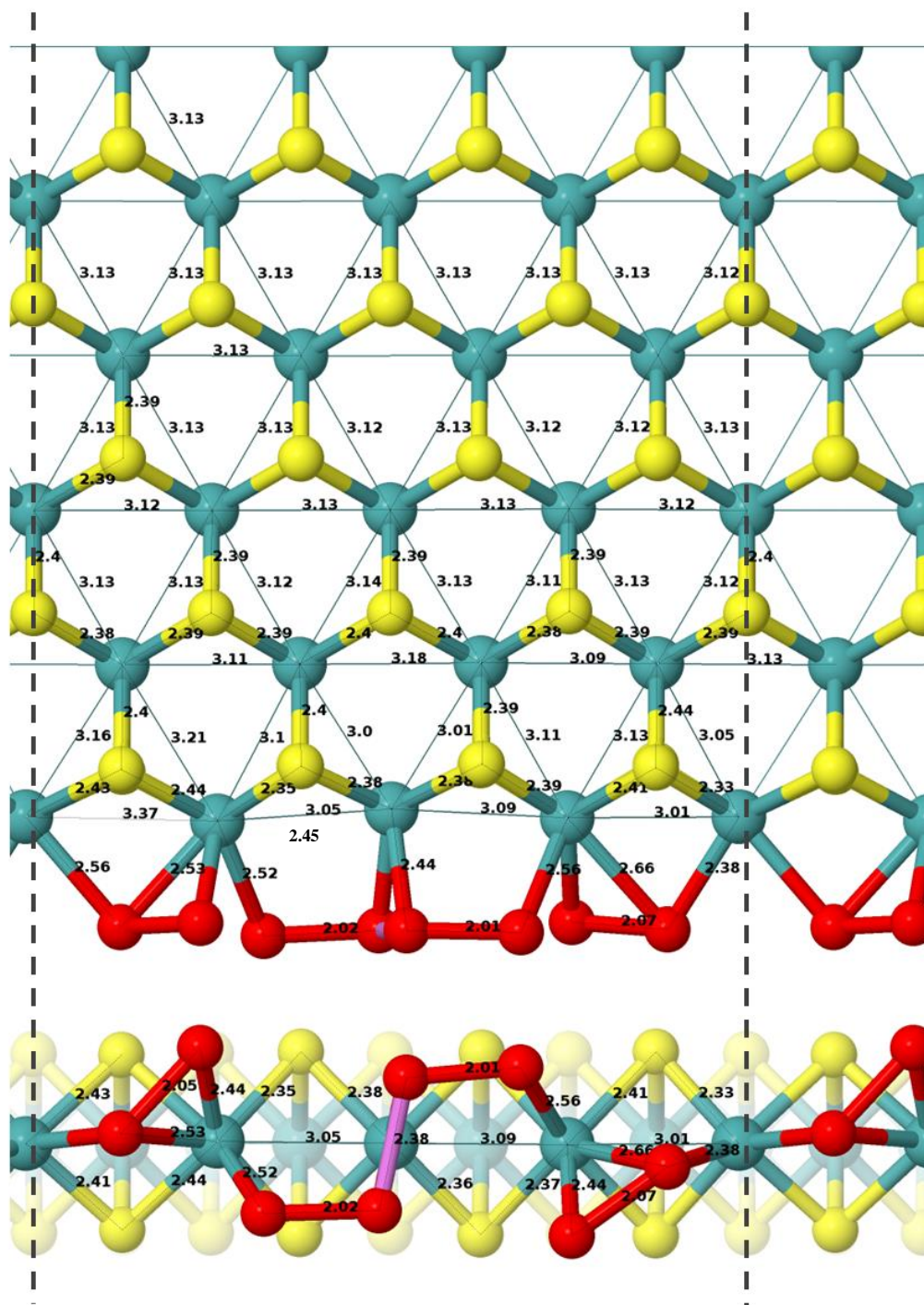


Figure S6 Top and side views of Mo-100%S (R3). Grey dashed lines indicate the length of the simulation cell. Atomic distances are in Angstroms. Mo-Mo and Mo-S bulk values are 3.13 Å and 2.39 Å respectively. Atom colour code: blue-green (Mo), yellow (S), red (edge S).

Mo-50%S configurations (Figure S7 & Table S1)

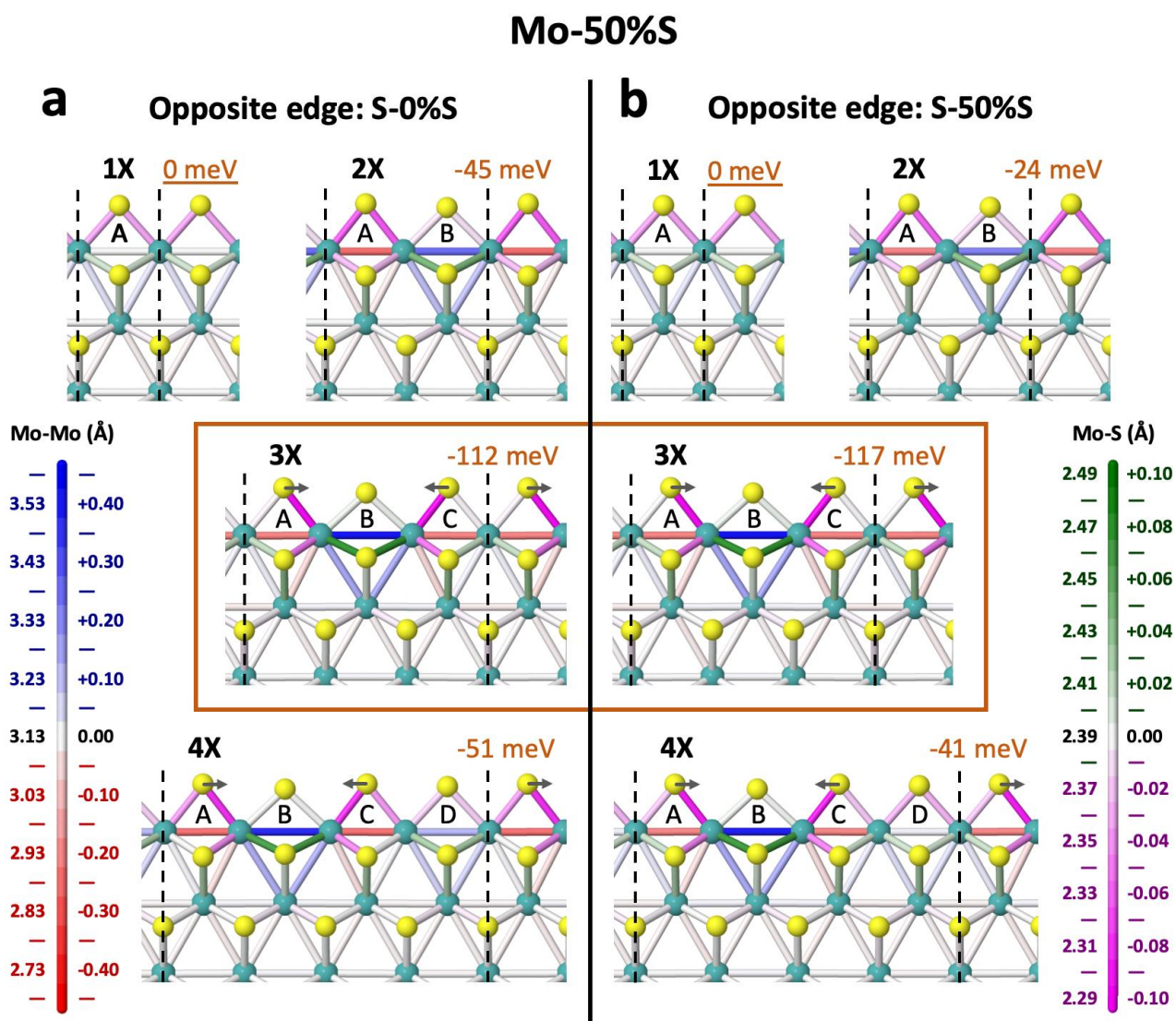


Figure S7 Top view of the Mo-50%S edge optimised structures for the different ribbon models presenting S-0%S (**a**) and S-50%S (**b**) as the opposite edge. For each supercell size ($L=1, 2, 3$ and 4) a different structure, matching the periodicity of the supercell, is found and is labelled as 1X, 2X, 3X and 4X respectively. The size of the unit cell is indicated by dashed lines. The bonds are coloured according to their length; with red (magenta) and blue (green) representing compressed and elongated Mo-Mo (Mo-S) bonds with respect to the bond distances in the infinite single-layer model (white). A colour key is presented for both Mo-Mo and Mo-S bonds with the absolute and relative distances indicated in the left and right part of each key respectively. Asymmetric S-bridges are indicated with arrows situated on the S atoms pointing to the edge direction for which they are displaced with respect to a symmetric situation. The relative energy per f.e.u. with respect to the 1X models for each type of ribbon (**a** and **b**), is indicated. In both cases the most stable model (enclosed in a brown box) is the one with 3X periodicity.

Supporting Information

Table S1 Bond angles and distances of the optimised Mo-50%S edge models composed of reconstructed S-bridges (S_B), for the structures obtained using cells of different length (L). The angles and distances presented correspond to the first row of S and Mo atoms at the edge: $-(\text{Mo-S})_L$. The bridges involved in the geometry parameters are specified with labels A-D as indicated in Figure S7. Values expressed without (with) parenthesis refer to models presenting in the opposite side of the ribbon the S-0%S (S-50%S edge). Bond distances (Å) and angles (°) are indicated by two and three atomic element symbols connected respectively.

L	S_B	Mo-Mo	Mo-S	Mo-S-Mo	S_B-S_B	S-S	$S_B-S_B-S_B$	S-S-S
1	A	3.127 (3.127)	2.360 (2.359)	83.0 (83.0)	A-A	3.127 (3.127)	A-A-A	180.0 (180.0)
2	A (out)	2.884 (2.923)	2.331 (2.334)	76.4 (77.6)	A-B	3.130 (3.129)	A-B-A	174.7 (175.7)
	B (in)	3.370 (3.331)	2.385 (2.380, 2.381)	89.9 (88.8)	B-A	3.130 (3.129)	B-A-B	174.7 (175.7)
3	A (out)	2.926 (2.924)	2.298, 2.389 (2.298, 2.391)	77.2 (77.1)	A-B	3.175 (3.175)	A-B-C	172.9 (172.8)
	B (in)	3.530 (3.533)	2.397 (2.397)	94.8 (95.0)	B-C	3.175 (3.175)	B-C-A	176.4 (176.4)
	C (out)	2.926 (2.924)	2.298, 2.389 (2.298, 2.391)	77.2 (77.1)	C-A	3.043 (3.043)	C-A-B	176.4 (176.4)
4	A (out)	2.890 (2.930)	2.356, 2.313 (2.370, 2.310)	76.5 (75.5)	A-B	3.164 (3.180)	A-B-C	173.0 (173.0)
	B (in)	3.492 (3.510)	2.394 (2.390)	93.7 (94.4)	B-C	3.165 (3.180)	B-C-D	174.7 (175.4)
	C (out)	2.890 (2.930)	2.356, 2.313 (2.370, 2.310)	76.5 (75.4)	C-D	3.097 (3.080)	C-D-A	176.5 (177.8)
	D (in)	3.236 (3.140)	2.376 (2.370)	85.8 (83.1)	D-A	3.096 (3.080)	D-A-B	174.7 (175.4)

Ribbon models (Figure S-8)

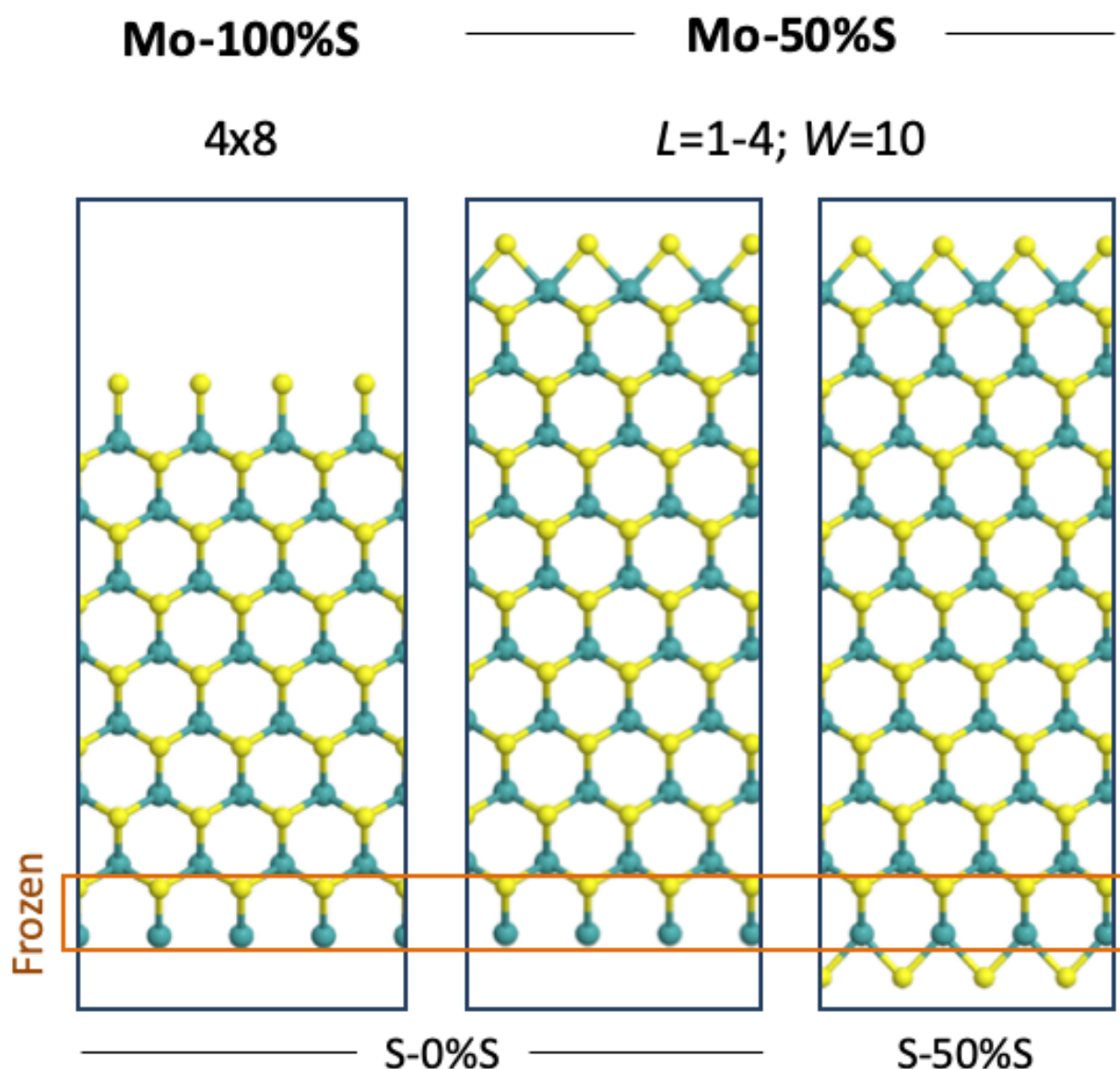


Figure S8 Top views of representative examples of complete ribbon models used in this work for the study of Mo-100%S and Mo-50%S edges. All Mo-100%S models have been studied using 4x8 ribbon presenting an opposite edge with a S-0%S configuration frozen in the bulk atomic positions. For the models with a Mo-50%S edge ribbon models of $W=10$ with different cell sizes ($L=1-4$) have been used. In this case both S-0%S and S-50%S configurations have been used as opposite edge. Atoms enclosed in an orange box have been kept frozen during the geometry optimisation at the bulk positions. Atom colour code: blue-green (Mo), yellow (S), red (edge S).