Electronic Supplementary Information (ESI)

A new twinning mechanism controlled by solute electronic structures

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Fig. S1. HAADF-STEM image of the W₂C phase and the region where EELS spectra were acquired are shown. a A low-magnification HAADF image of the W₂C phase highlights EELS collection region, marked by a yellow box. **b** The high-resolution HAADF image displays the atomic-scale structure of the W₂C phase.



Fig. S2. Atomic-resolution HAADF images and intensity profiles across the (V, W)C-WC interface with distinct stacking sequences. a-c Stacking sequences of the (V, W)C regions (*ABACBA*, *ABCBCA*, and *ABCABC* sequences, respectively, from bottom layer to top layer) are shown, overlaid with atomic models to highlight structural differences. d-f Corresponding HAADF intensity profiles extracted from the boxed regions, vertically integrated along the P1 \rightarrow P2, P3 \rightarrow P4, and P5 \rightarrow P6 lines.

Layer	3	2	1	0	ī	2	3	4	5
V	4.7	6.5	21.6	57.6	45.6	54.5	63.7	65.8	69.3
Error	0.8	1.0	2.2	3.0	3.1	3.0	2.8	2.8	2.6
W	95.3	93.5	78.4	42.4	54.4	45.5	36.3	34.2	30.7
Error	0.8	1.0	2.2	3.0	3.1	3.0	2.8	2.8	2.6

 Tables S1 Measured chemical compositions of different atomic layers from the P1 in WC side to the P2 in (V, W)C side.

Tables S2 Measured chemical compositions of different atomic layers from the P3 in WC side to the P4 in (V, W)C side.

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Layer	3	2	1	0	ī	2	3	4	5
V	3.4	8.7	20.4	59.0	59.3	49.5	50.9	66.1	66.8
Error	0.8	1.4	2.1	2.9	2.9	3.1	3.1	2.7	2.7
W	96.6	91.3	79.6	41.0	40.7	50.5	49.1	33.9	34.2
Error	0.8	1.4	2.1	2.9	2.9	3.1	3.1	2.7	2.7

 Tables S3 Measured chemical compositions of different atomic layers from the P5 in WC side to the P6 in (V, W)C side.

Layer	3	2	1	0	ī	2	3	4	5
V	6.1	5.9	13.1	57.6	81.8	86.3	88.3	86.9	87.9
Error	1.0	1.3	2.0	3.0	1.9	1.5	1.3	1.4	1.4
W	93.9	94.1	86.9	42.4	18.2	13.7	11.7	13.1	12.1
Error	1.0	1.3	2.0	3.0	1.9	1.5	1.3	1.4	1.4