

Figure 1 | Multi-junction lateral heterostructures and interfaces based on MoSe₂ and WSe₂. a, Low-magnification optical image of three-junction heterostructures. The inset shows a larger magnification of the area within the dashed box. The dark-contrast regions correspond to MoSe2, the bright-contrast regions to WSe₂, **b**, **c**, Optical images of five-junction heterostructures. The difference in thickness of the MoSe₂ layers in **b** and **c** is seen by the difference in thickness of the dark-contrast regions. d, Seven-junction heterostructure with variable domain widths. The underlying colour bars in **b-d** depict the growth timescale: from left to right (pink, MoSe2; green, WSe2), each division (black line) corresponds to approximately 120 s. e, f, Raman (e) and photoluminescence (PL, f) spectra, of a at positions 1, 2, 3 and 4. g, h, Photoluminescence intensity maps for the WSe₂ $(1.6 \,\mathrm{eV}, \mathrm{top})$ and MoSe₂ $(1.52 \,\mathrm{eV},$ bottom) domains (g), and composite photoluminescence map (h) for the heterostructure in b. i, j, Contour colour plots of the normalized photoluminescence intensity of three-junction (i) and fivejunction (j) heterostructures, along the arrows in the insets. k, l, Z-contrast atomicresolution HAADF-STEM images of pure MoSe₂ (k) and WSe₂ (l). m, n, Atomic resolution HAADF-STEM images of the smooth (m) and sharp (n) interfaces, with their corresponding Fourier-transform patterns and composition profiles (atomic fraction of tungsten per vertical atomic column). The smooth and the sharp interfaces have average interface widths of 6 nm (21 atomic columns) and 1 nm (4 atomic columns), respectively. o, p, Scattered electron intensity colour plot (o) and associated atomic ball model (p) for the junction in n. q, Electron intensity profile along the white box in i. Scale bars, $10 \mu m$ (a (inset), $\mathbf{b} - \mathbf{d}$, \mathbf{g} , \mathbf{h}).

change in the position of the photoluminescence peak, suggesting the formation of sharper interfaces with less alloying. At these sharp interfaces, the photoluminescence spectra are characterized by an overlap of both peaks; this is due to the submicrometre laser spot size in the confocal microscope simultaneously probing both sides of the interface. Although junctions 2 and 3 are both sharper than junction 1, it is worth noting that junction 3 is not as sharp as junction 2; this behaviour has been consistently observed in all samples. It indicates that a transition from a MoSe₂ to a WSe₂ domain results in a less abrupt, slightly 'smoother' interface between the two materials, whereas the transition from a WSe₂ to a MoSe₂ domain produces atomically sharp interfaces. This was verified by atomic-resolution Z-contrast imaging using high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) (Fig. 1k-o), which provides insight into both the crystalline quality and the chemical distribution at heterojunctions at a high spatial resolution. The atoms in monolayer MoSe₂ (Fig. 1k) and WSe2 (Fig. 1l) have a hexagonal arrangement (honeycomb-like) with D_{3h} symmetry. The atomic positions of both Mo and Se2 yield a similar intensity of scattered electrons, whereas the W sites display twice that intensity (Fig. 1q)¹². Figure 1k, l shows pure MoSe₂ and WSe₂ regions, respectively, within the same heterostructure, confirming that the evaporation-deposition process is very selective even though both solid precursors (MSe2 and WSe2) are present in the heterogeneous source. Consistent with the photoluminescence

observations, two types of interface were identified: $MoSe_2 \rightarrow WSe_2$ interfaces (Fig. 1m), which display a smooth, less abrupt chemical transition with some degree of alloy formation, and $WSe_2 \rightarrow MoSe_2$ interfaces, which are atomically sharp (Fig. 1n).

The different interfaces are a consequence of the different oxidation and reduction rates of molybdenum and tungsten compounds²² as well as the gas switching mechanism. When the carrier gas switches from H₂O to H₂, the residual metal oxide content depletes rapidly. Because the complete replacement of H₂O to H₂ is not possible in the present experimental setup, this results in a small amount of co-deposition of Mo in the WX₂ domain, hence forming a smooth interface (MoX₂ \rightarrow WX₂). Under H₂ flow, while the WX₂ domain continues to grow, the molybdenum oxide(s) are converted completely to metallic molybdenum over the MoX₂ source. When the conditions are reversed—switching from H₂ to H₂O vapour again—the low-index W sub-oxides begin to form high-index W sub-oxides, as indicated by the slow weightloss rate of the tungsten oxide precursor in H₂O (Extended Data Fig. 5c, d). Meanwhile, H₂O restores the initial oxidation step from metallic molybdenum—formed over MoX2 during interaction with H2 gas—to MoO₂, which is relatively slower than the direct oxidation of the MoX₂ source. This might lead to a delayed supply of MoO₂ vapour to the already present WX₂ edge-site, and hence result in a sharp transition from the WX₂ to the MoX₂ domain. Further optimization of the gas switching process could lead to the generation of sharp interfaces only.