

Supplementary Pages (Optional)

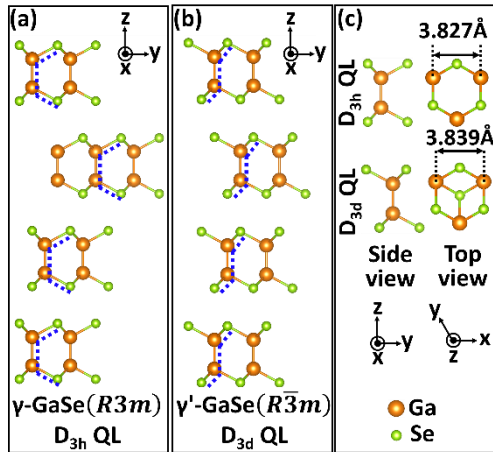


Fig. S1 Schematic models of (a) γ -GaSe, (b) γ' -GaSe, (c) side (left) and top (right) views of two GaSe quadruple layers (QLs) polymorphs, D_{3h} (axial symmetry, top) and D_{3d} (central symmetry, bottom). The blue dashed lines highlight the stacking configuration. The thickness of GaSe QL is about 8 Å. The in-plane lattice parameters of D_{3h} and D_{3d} GaSe QL are 3.827 Å and 3.839 Å, respectively.

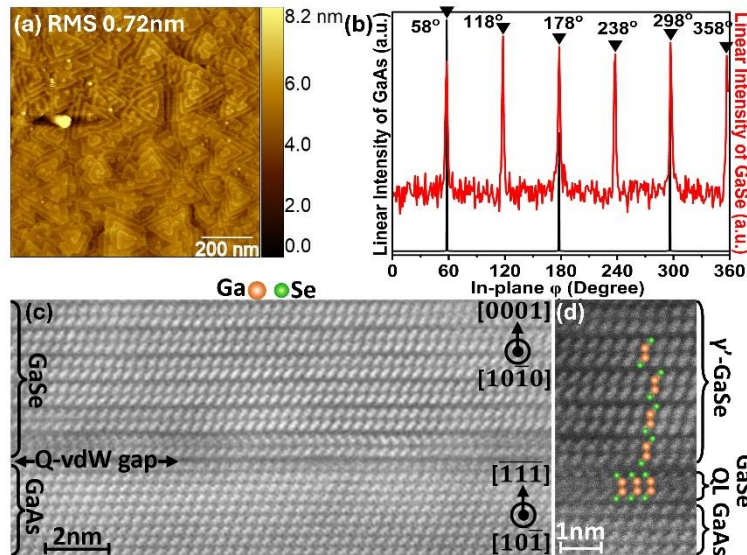


Fig. S2 (a) Atomic force microscopy image of a GaSe sample. RMS is the root-mean-square surface roughness. (b) In-plane ϕ scans of a GaSe sample. “▼” and “▽” designate the main and minor diffraction peaks of GaSe, respectively. The in-plane scans of the GaSe film and GaAs substrate were around the (103) plane and (311) planes, respectively. The out-of-plane orientations for GaSe and GaAs were (002) and (111), respectively. Cross-sectional annular dark field-scanning transmission electron microscopy images of a GaSe sample at (c) low magnification (low-pass filtered to reduce noise) and (d) high magnification.

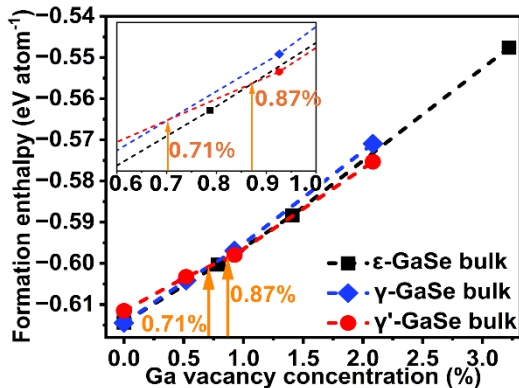


Fig. S3 GaSe formation enthalpy as a function of Ga vacancy concentration in bulk GaSe crystals. Inset is a magnified view.