Free-Standing Large, Ultrathin Germanium Selenide van der Waals Ribbons by

Combined Vapor-Liquid-Solid Growth and Edge Attachment

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Supporting Figures





Figure S1. Morphology of GeSe nanoribbons. **(a)** SEM of Au-catalyzed GeSe nanoribbons on the Au/Si growth substrate. **(b)** Higher magnification SEM image of the GeSe ribbons. **(c)** Trends of thickness, base width, and length of the GeSe ribbons as a function of growth temperature.



Figure S2. (a) SEM of Au-catalyzed GeSe nanoribbons on the Si growth substrate. **(b)** EDS chemical maps showing the distribution of Ge and Se on the growth substrate.



Figure S3. (a) SEM of Au-catalyzed GeSe nanoribbons transferred to SiN_x/Si support. (b) – (d) EDS chemical maps showing the distribution of Ge, Se in the nanoribbons and Si stemming from the support.



Figure S4. Electron microscopy and diffraction of GeSe ribbons. (a) TEM image of characteristic GeSe nanoribbons on amorphous carbon film (Growth temperature: 320°C). **(b), (c)** Nanobeam electron diffraction of the two nanoribbons in (a). **(d)** High-resolution TEM image of a GeSe ribbon at the position marked with the white box in (a).



Figure S5. Morphology of GeSe ribbons. (a) TEM image of a characteristic GeSe nanoribbon on amorphous carbon film (Growth temperature: 340°C, growth substrate Au/Si). Inset: Nanobeam electron diffraction of of the ribbon along the [001] zone axis. **(b)** HAADF-STEM image of the nanoribbon in (a). **(c)** Higher magnification TEM image of the tip of the GeSe ribbon.



Figure S6. Morphology of GeSe ribbons grown with Au catalyst on graphite. (a) TEM image of a characteristic GeSe nanoribbon on amorphous carbon film (Growth temperature: 340°C, growth substrate Au/graphite). Inset: Nanobeam electron diffraction of the ribbon along the [001] zone axis. **(b)** HAADF-STEM image of the nanoribbon in (a).



Figure S7. Morphology of GeSe ribbons grown with Au catalyst on graphite. (a) TEM image of a characteristic GeSe nanoribbon on amorphous carbon film (Growth temperature: 340°C, growth substrate Au/graphite). Inset: Nanobeam electron diffraction of the ribbon viewed along the [001] zone axis. **(b)** HAADF-STEM image of the nanoribbon in (a).



Figure S8. Raman spectroscopy of GeSe nanoribbons. (a) Optical microscopy image of a GeSe nanoribbon grown at 320°C. **(b)** – **(c)** Micro-Raman intensity maps of the A_g and B_{3g} Raman modes of GeSe.



—— 5 μm

Figure S9. Analysis of the tapered shape of GeSe nanoribbons. (a) Optical microscopy image of a GeSe nanoribbon grown at 320°C (transferred to SiN_x chip). **(b)** Measurements of the half-width at the base (*d*) and length (*L*) of the ribbon segment, along with the opening angle (θ). Here, $d = 2.3 \ \mu\text{m}$ and $L = 33.3 \ \mu\text{m}$, so that $\theta = 2 \cdot \arctan(d/L) = 7.9^\circ$. **(c)** Histogram of the distribution of opening angles θ for an ensemble of 30 nanoribbons supported on a flat SiN_x chip. **(d)** Histogram of the ratio between length and base half-width, showing $L:d = (11.5 \pm 1.8)$.



Figure S10. Cathodoluminescence spectroscopy of individual GeSe nanoribbons. (a) HAADF-STEM image of a typical GeSe nanoribbon. **(b)** Panchromatic CL map of the GeSe nanoribbon.



Figure S11. (a) – **(b)** Transmission electron microscopy images of Au film deposited on graphite by sputtering, following dewetting at the growth temperature of 340°C.



Figure S12. Transmission electron microscopy image of a typical GeSe flake grown on Si substrate without deposited Au at a growth temperature of 340°C. The flake, transferred to lacey carbon film for the TEM imaging, shows a typical irregular shape with a long, straight edge that likely represents its interface where it broke off the Si growth substrate.