SUPPLEMENTARY INFORMATION for

Screw-Dislocation-Driven Growth Mode in Two Dimensional GaSe on GaAs (001) Substrates grown by Molecular Beam Epitaxy

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Figure S1. Sketch of the growth process of without (a) and with (b) Se-pretreated samples

Figure S2. In-situ RHEED patterns: (a) and (b) represented to GaAs(001) substrate after thermal oxide desorption; (c) and (d) represented to non Se-pretreated sample during the materials growth; (e) the intensity profiles extracted from (c) and (d); and (f) the sketch of atomic alignment between GaSe layer and GaAs(001).



Figure S3. AFM images, Raman scattering, and photoluminescence spectrum of GaSe grown on GaN/Sapphire substrate



Figure S4. XRD 2 θ -scanning spectra of GaSe sample C in comparison to GaSe bulk. The inset shows the 2 θ -scanning spectra of (004)-peak with the step scan size of 14 arcsec.



Figure S5. Raman spectrum of sample C compare to that of the bulk under 514 nm wavelength excitation.



Figure S6. In-situ RHEED patterns: (a) and (b) represented to Se-pretreated GaAs(001) substrate; (c), (d) represented to Se-pretreated sample C-Se during the material growth; (e) the sketch of atomic alignment between GaSe layer and the Se-passivated GaAs(001) substrate.



In comparison to the non Se-pretreated GaAs surface (Fig. S2a), the RHEED pattern of the Sepretreated GaAs surface (Fig. S6a) shows extra lines in between spotty lines of GaAs. It is attributed to the presence of Se on GaAs surface.