

Supplementary Information for

Understanding the Growth Mechanisms of γ -GeSe for Polymorph-Selective Large-Area Growth

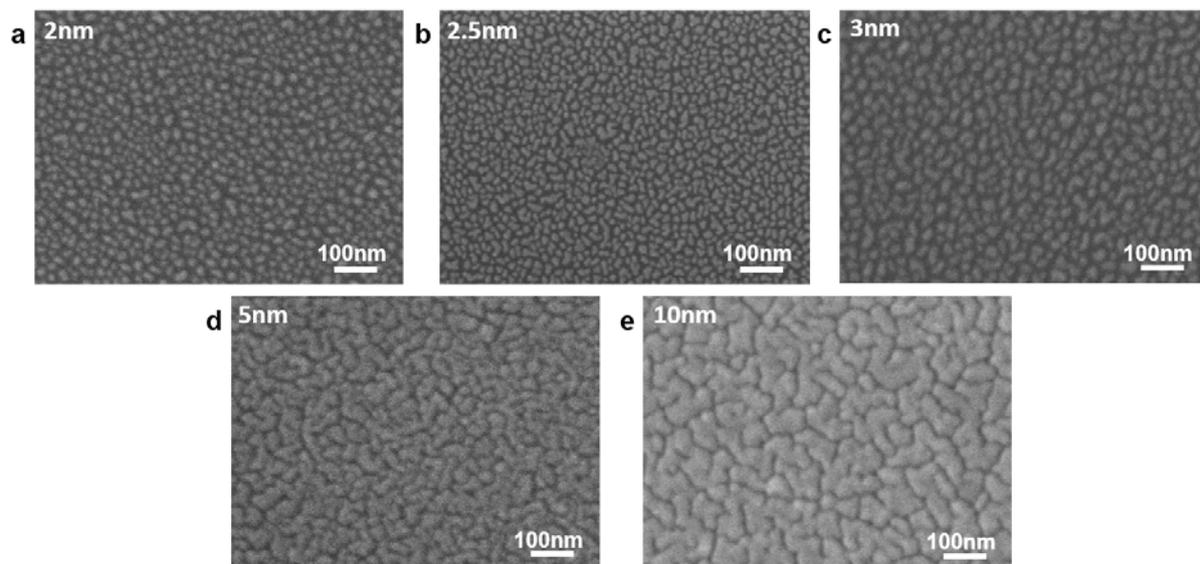
*Joong-Eon Jung,¹ Sol Lee,¹ Hani Kang,¹ Myeongjin Jang,¹ Jinsub Park,¹ Petri Mustonen,²
Harri Lipsanen,² Zhipei Sun,² Hoon Hahn Yoon³ and Kwanpyo Kim^{1,*}*

¹Department of Physics, Yonsei University, Seoul 03722, Korea

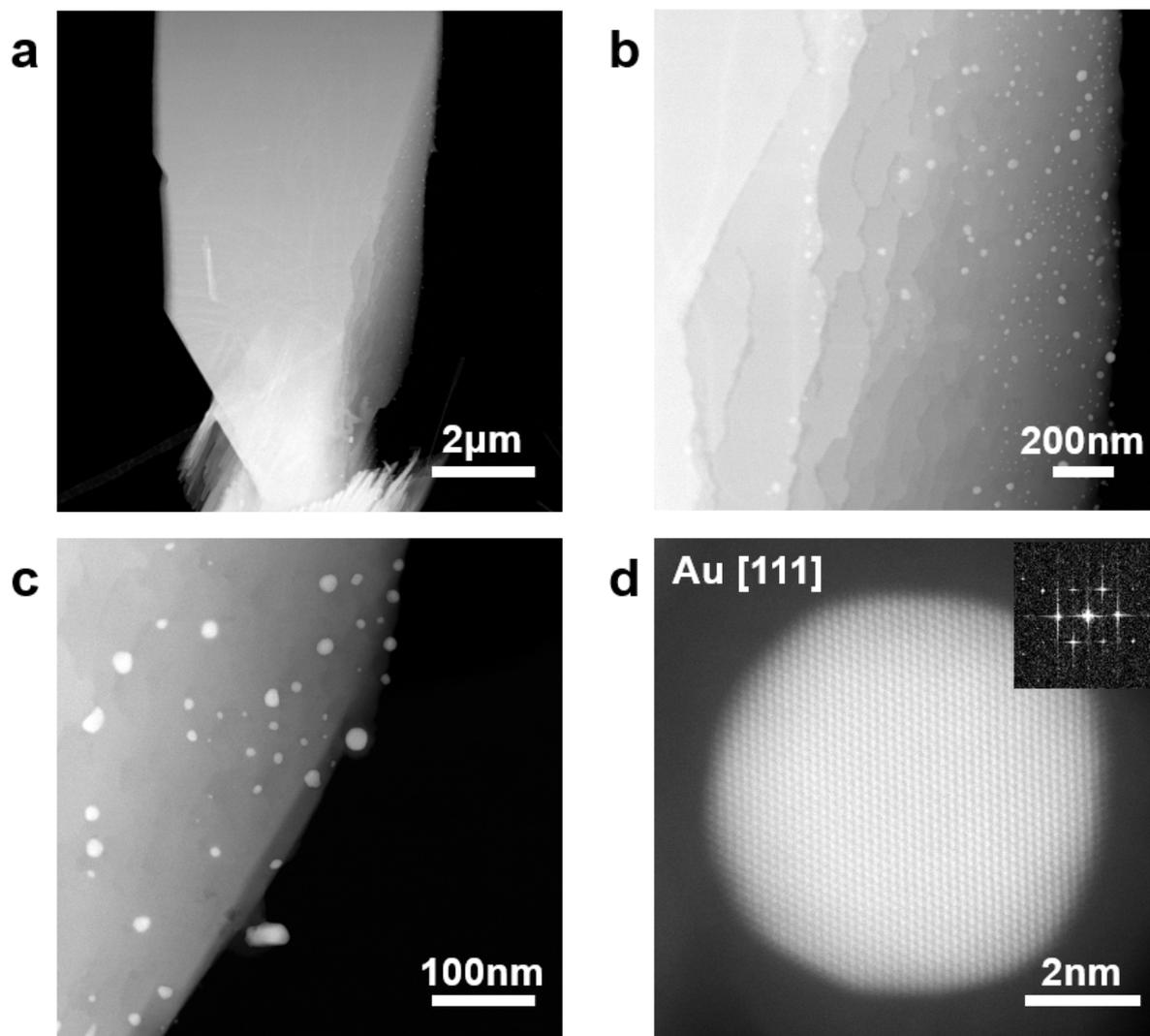
²Department of Electronics and Nanoengineering, Aalto University, Espoo 02150, Finland

³Department of Semiconductor Engineering, School of Electrical Engineering and Computer Science, Gwangju Institute of Science and Technology, Gwangju, 61005, Republic of Korea

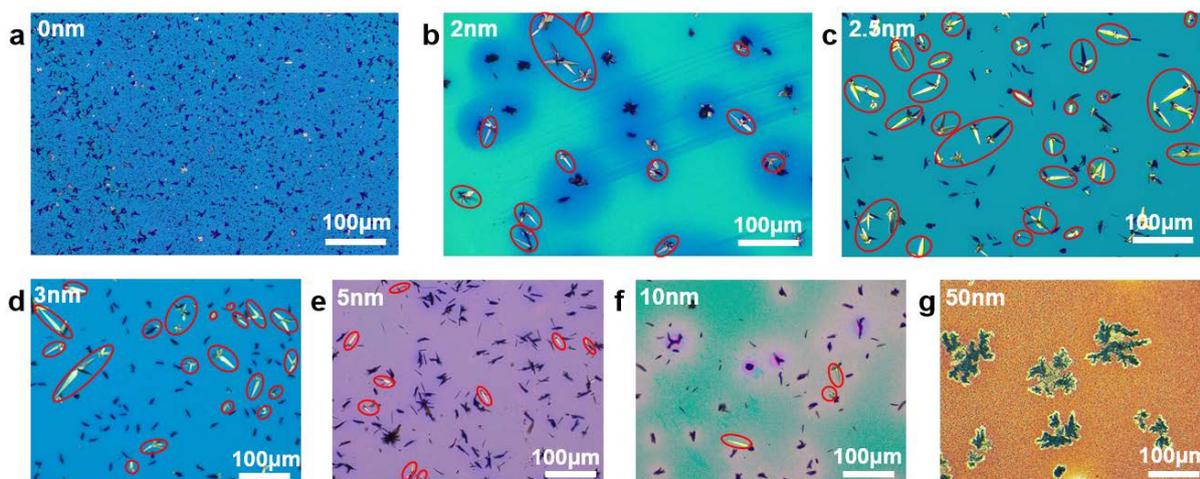
*Address correspondence to K.K. (kpkim@yonsei.ac.kr)



Supplementary Figure S1. SEM images of Au deposited at various thicknesses on SiO₂/Si substrates. (a) Au 2 nm, (b) 2.5 nm, (c) 3 nm, (d) 5 nm, and (e) 10 nm.

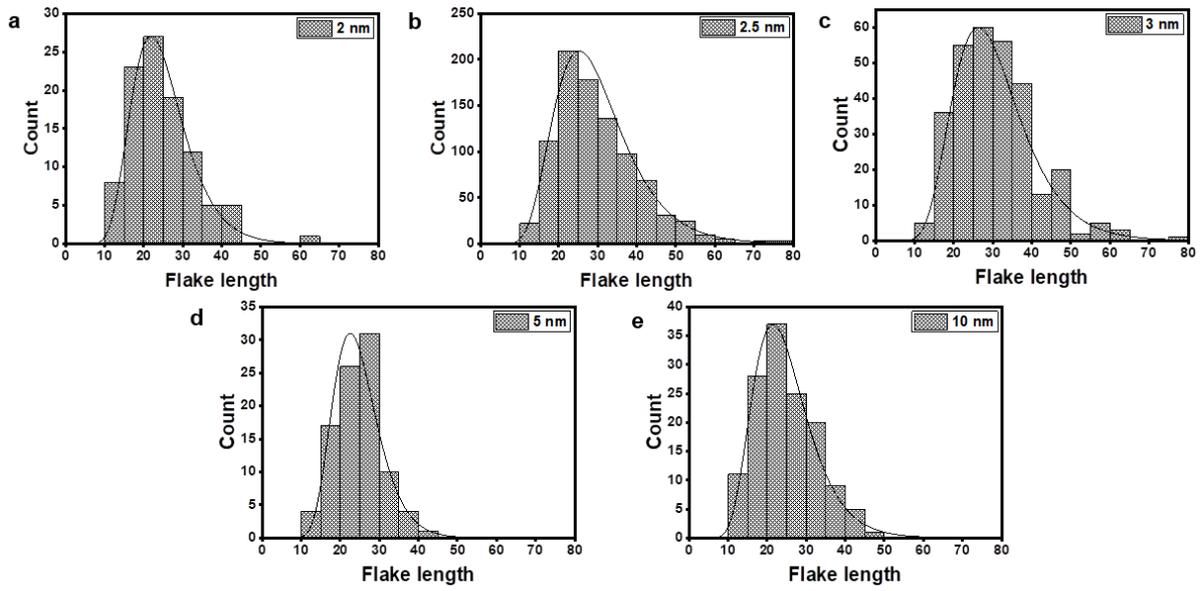


Supplementary Figure S2. Au nanoparticles on the surface of γ -GeSe. (a) STEM image of a γ -GeSe flake. (b,c) Enlarged images from panel a. Multiple Au nanoparticles on the γ -GeSe surface were observed. (d) Zoomed-in STEM image of an Au nanoparticle with [111] zone axis. The inset shows the fast Fourier transform (FFT) image.

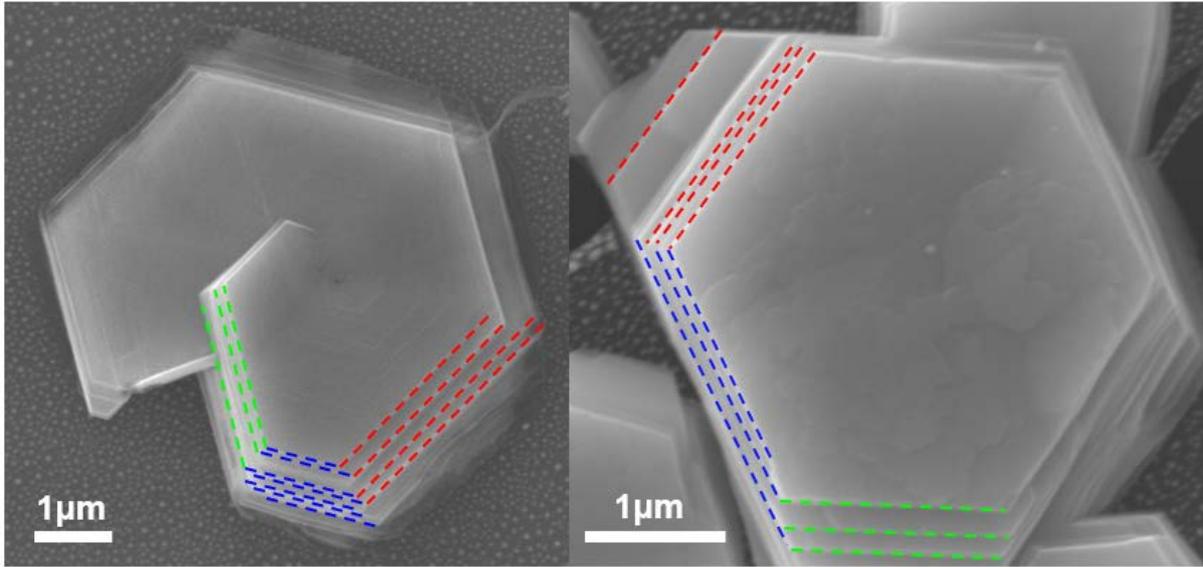


Supplementary Figure S3. γ -GeSe synthesis depending on the thickness of deposited Au.

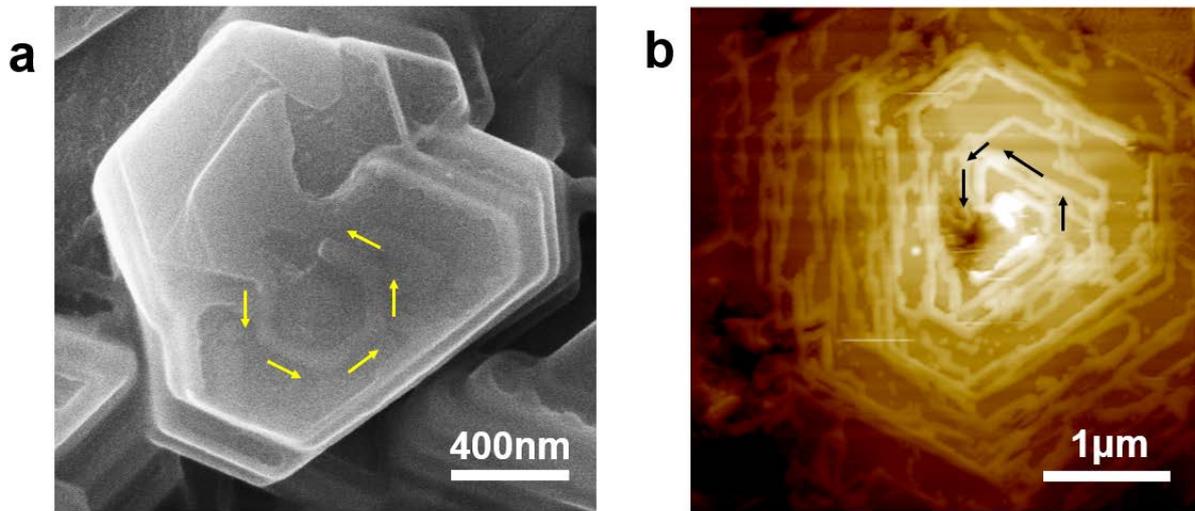
(a) Optical image of growth product without Au deposition. (b) Optical image of synthesizing γ -GeSe by depositing 2 nm of Au. (c) 2.5 nm. (d) 3 nm. (e) 5 nm. (f) 10 nm. (g) Optical image of growth product with 50 nm of Au. γ -GeSe was not synthesized. The crystals marked with red circles are γ -GeSe.



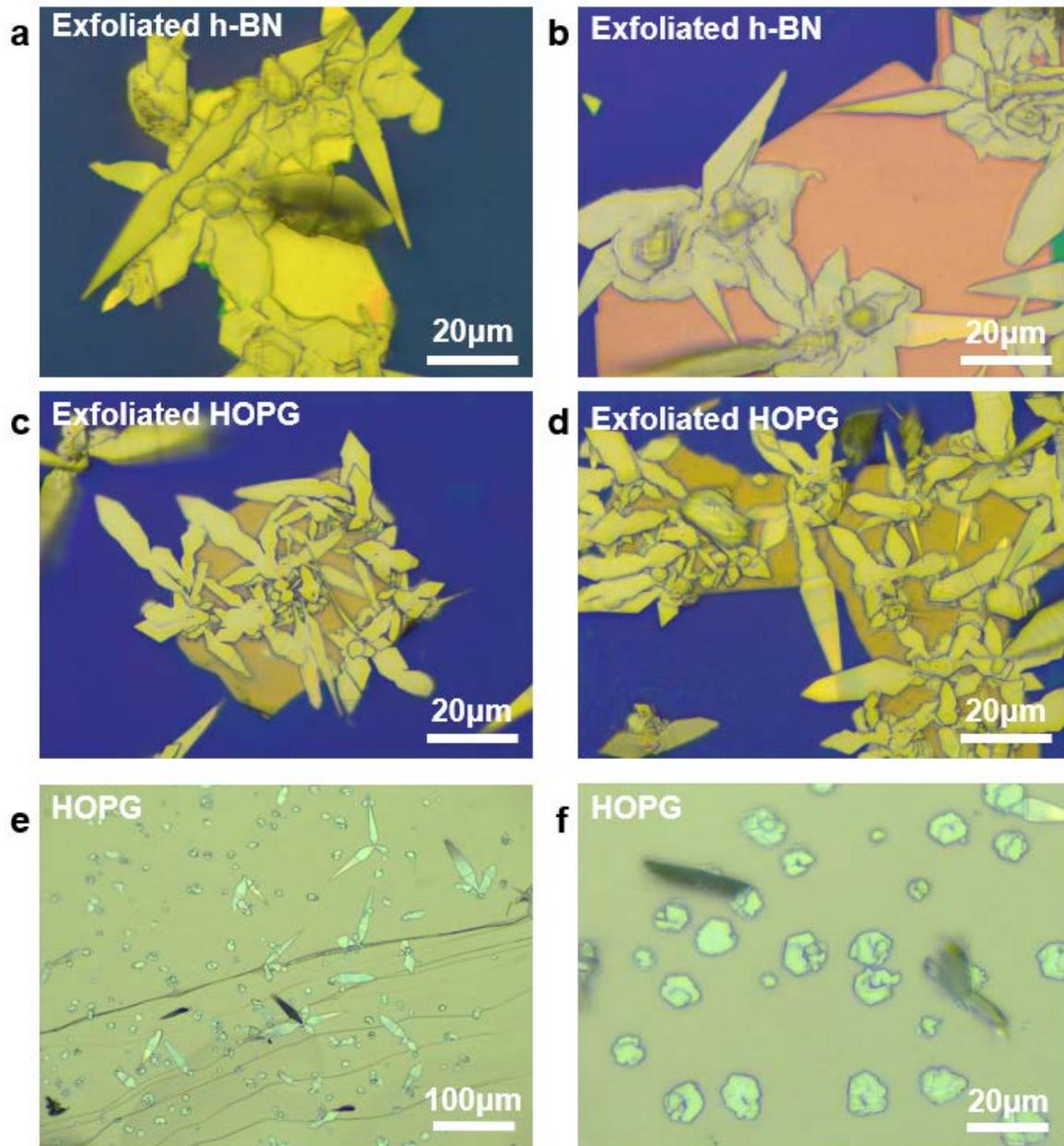
Supplementary Figure S4. Histogram of γ -GeSe flake length depending on the thickness of deposited Au. (a) 2 nm, (b) 2.5 nm, (c) 3 nm, (d) 5 nm, and (e) 10 nm.



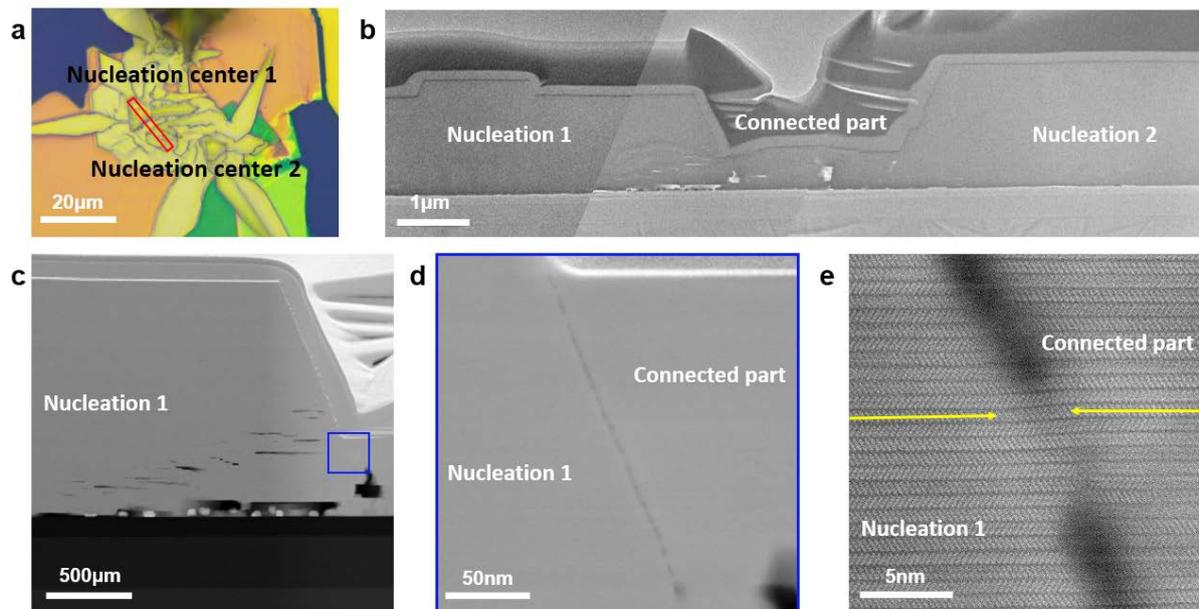
Supplementary Figure S5. Spiral growth of γ -GeSe crystal. SEM images of γ -GeSe display the spiral growth shape. Dashed lines indicate the crystal edges.



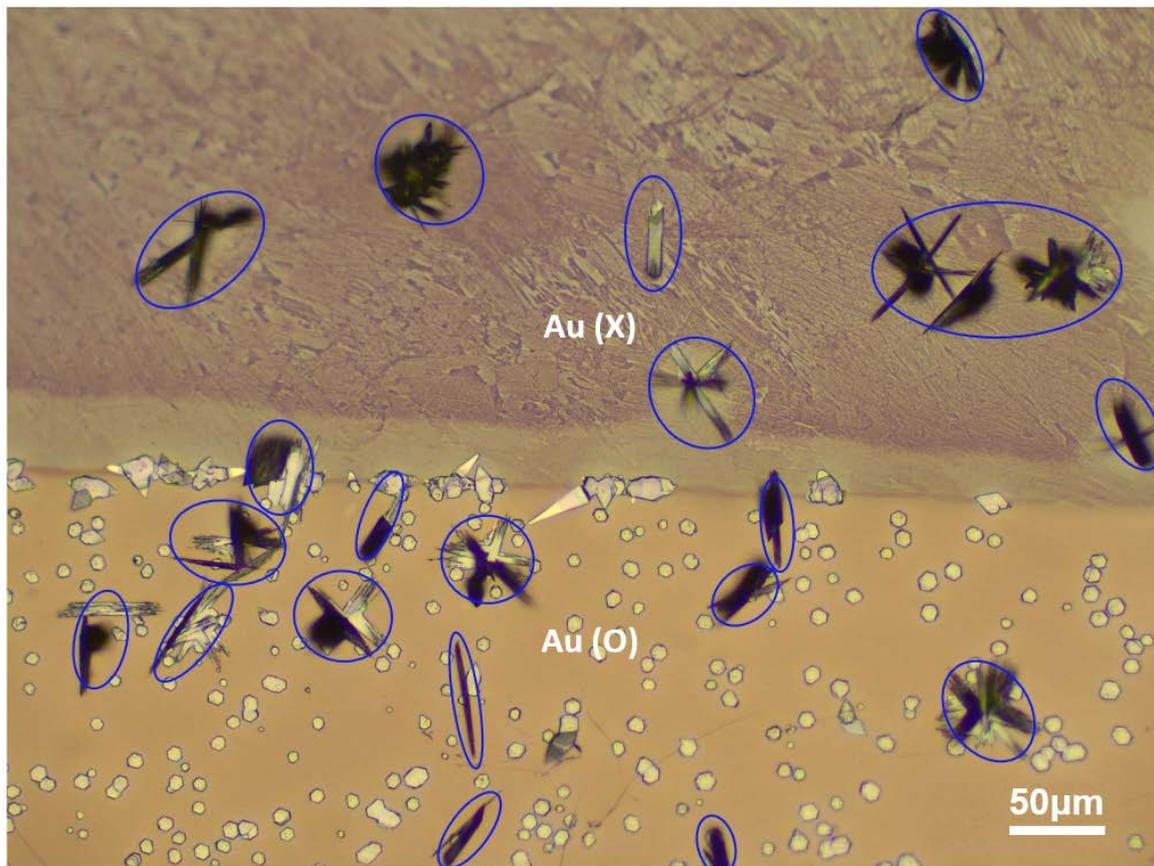
Supplementary Figure S6. Investigation of γ -GeSe surface topography. (a) SEM image of the alloy droplet trajectory on the γ -GeSe surface. Yellow arrows indicate the trajectory of droplet movement. (b) AFM measurement of γ -GeSe crystal. Black arrows indicate the trajectories.



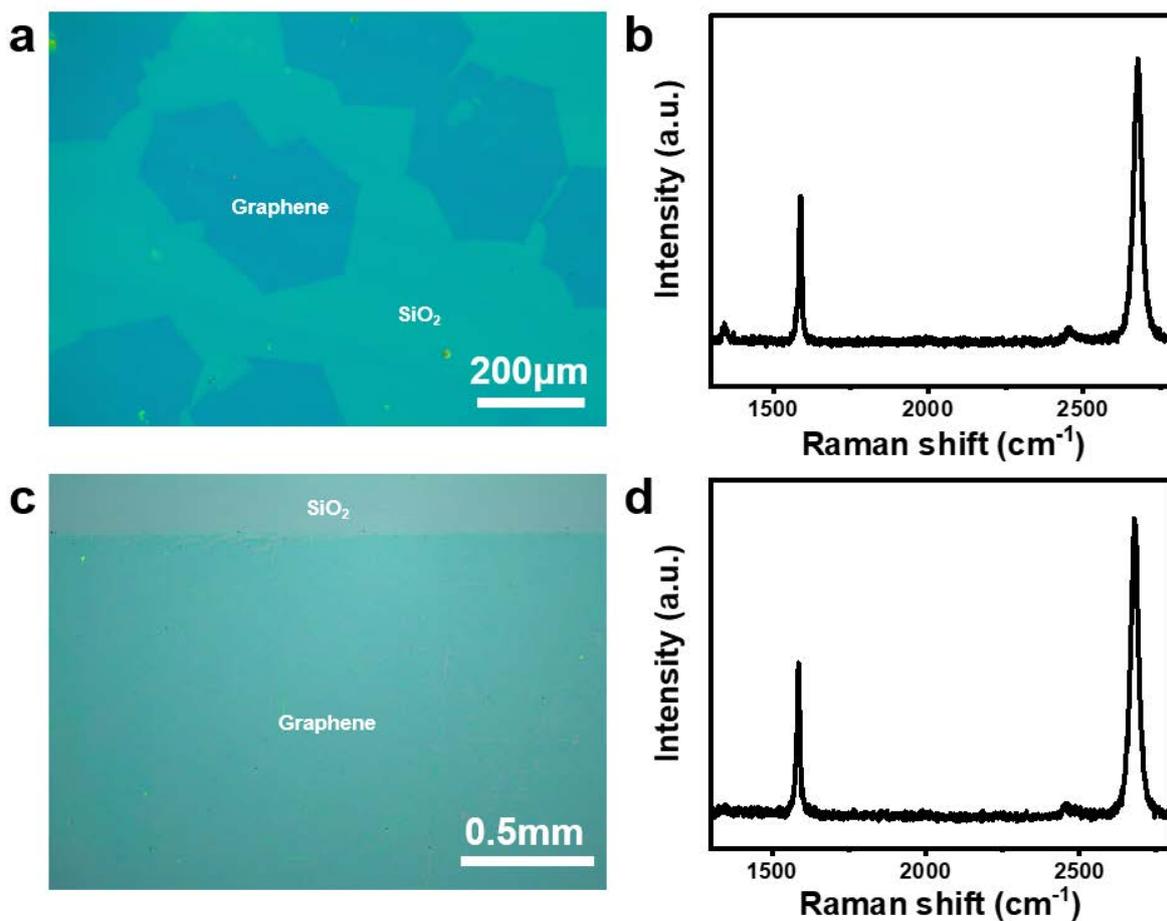
Supplementary Figure S7. γ -GeSe grown on h-BN and HOPG. (a,b) Optical images of γ -GeSe grown on exfoliated h-BN. The flakes grown from different seeds are connected. (c,d) Optical images of γ -GeSe grown on exfoliated HOPG. (e,f) Optical images of γ -GeSe grown on HOPG substrates.



Supplementary Figure S8. Investigation of γ -GeSe crystal connection grown on h-BN substrate. (a) Optical image of connected γ -GeSe grown from different locations on h-BN. (b) Cross-sectioned TEM image of the area in the red box. (c) Magnified STEM image of nucleation 1 region. (d) Magnified STEM image of the area in the blue box of panel c. (e) STEM image showing the connection between nucleation 1 and the connected part. The yellow arrow shows the connection of the atomic layers of γ -GeSe.



Supplementary Figure S9. Synthesis of GeSe with and without Au on HOPG substrate. Optical image of γ -GeSe synthesized by depositing Au on only half-bottom of the HOPG substrate. γ -GeSe grows only below the boundary line. The crystals marked with blue circles are α -GeSe.



Supplementary Figure S10. Characterizations of CVD graphene. (a) Optical image of the single crystalline graphene on SiO₂/Si substrate. (b) Raman spectrum of the single crystalline graphene. (c) Optical image of the polycrystalline graphene on SiO₂/Si substrate (d) Raman spectrum of the polycrystalline graphene.

	a [Å]	Lattice mismatch with γ-GeSe $100 \times (3a_{\text{substrate}} - 2a_{\gamma\text{-GeSe}})/2a_{\gamma\text{-GeSe}}$
γ-GeSe	3.73	N. A.
h-BN	2.50	0.54 %
Graphite	2.46	-1.07 %

Supplementary Table S1. Lattice parameters of γ -GeSe and hexagonal substrates.