Supplementary material

1. Boat design

The LPE growth was realized through a slide boat system with a boat designed shown in Fig. S1. Four source wells were made for multi-layer deposition, and a thermocouple hole was for inserting the thermos-couple to measure the temperature where could be close to the actual growth location. Four little caterpillar wells are made beside the source wells to reduce the residue solution after growth using caterpillar effects. Shims with different thickness are also made and to be selected according to the substrate height in order to reduce the residue amount as much as possible. The precursors, namely Pb shots and Ge pieces will be put into the well with the well cap, and the well cap is designed to prevent the precursors from sliding out the well during installation.



Fig. S1. Schematics of the boat designed for LPE growth.

2. Proof of (111) facets

The cross-section SEM image of an LPE sample shows that for each of the LPE Ge grain, the side surfaces are around 54.7° from the bases. And this is valid for statistically for averaging 10 grains measured in one sample, and totally 3 samples were measured. And the angle between (111) facets and (001) base are exactly 54.7°.



Fig. S2. Cross-section SEM image of the LPE Ge over the Ge template.

3. Proof of epitaxy of LPE Ge layer over Ge (111) wafer

The θ -2 θ scan of the LPE Ge over Ge (111) wafer and the Ge (111) wafer itself is shown in Fig. S3. Both of the samples have the Ge (111) peak. No peaks at other orientations are found. The weak peak around 24.6° does not belong to Ge, which could be a secondary phase in the Ge wafer. The ϕ scans of the Ge (004) peak integrated from (111) pole figure for both the LPE Ge over Ge (111) wafer and the Ge (111) wafer are shown in Fig. S4. The ϕ spectrum of the Ge (111) wafer shows a three-fold symmetry, while that of the LPE Ge over the Ge (111) wafer has a six-fold symmetry, indicating that LPE Ge grows epitaxially over the Ge (111) wafer. The six-fold symmetry is probably caused by nucleation over stacking faults.



Fig. S3. HRXRD θ -2 θ scans of the LPE Ge over Ge (111) wafer and the Ge (111) wafer.



Fig. S4. ϕ scans of the Ge (004) peak integrated from the pole figure measurement of both the LPE Ge over Ge (111) wafer and the Ge (111) wafer.

4. Proof of strain in the substrate Ge

A T64000 Raman spectrometer equipped with a microscope with 100× objective was used to collect the Raman spectra with a 458 nm (Ar⁺) excitation source. The incident and the reflected beams are along the normal of the samples during the Raman measurement. The Raman patterns of the substrate Ge and a Ge (001) wafer are shown in Figure S5. The peak position of the Ge wafer is ~300.3 cm⁻¹, which is close to the value of bulk Ge¹ (300.8 cm⁻¹). A compressive strain in the substrate Ge film is indicated by the right shift of the Raman peak position to ~302.9 cm⁻¹, which supports the conclusion derived by the XRD 2 θ peak positions of the substrate Ge.



Figure S5. Raman spectra of the substrate Ge and a Ge (001) wafer.

References

1. H. N. Chong, Z. W. Wang, C. N. Chen, Z. M. Xu, K. Wu, L. Wu, B. Xu and H. Ye, Journal of Crystal Growth, 2018, 488, 8-15.