Supporting Information for

Growth and in-Plane Undulations of GaAs/Ge Superlattices on [001]-Oriented Ge and GaAs Substrates: Formation of Regular 3D Island-in-Network Nanostructures

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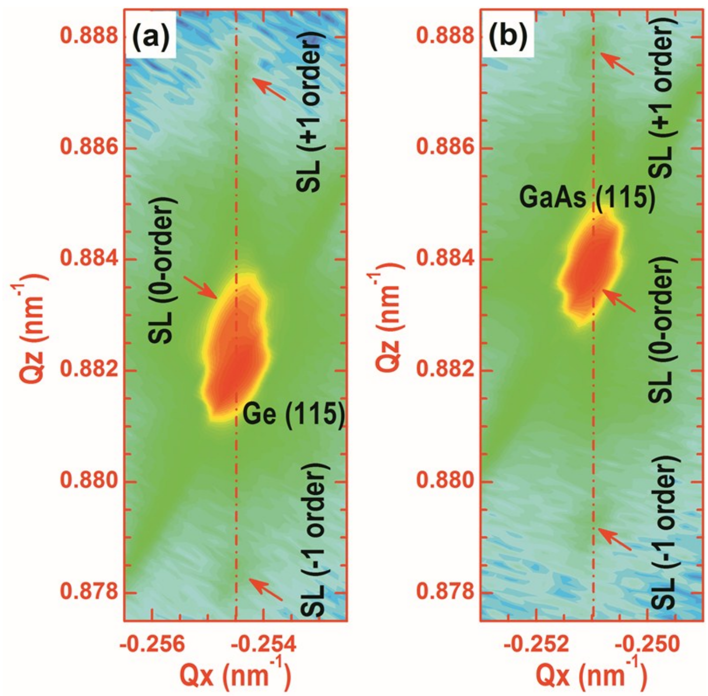

Figure S1 HRXRD mappings collected around the (115) atomic planes from typical 20-period GaAs/Ge superlattice structures grown by MOCVD on [001]-oriented Ge (a) and GaAs (b) substrates. The line connecting the ± 1st-order fringes and passing through the diffraction peaks

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of the substrates [i.e., the Ge (115) and GaAs (115) in (a) and (b), respectively] provide evidence that the superlattice structures are coherently strained on both the Ge and GaAs substrates.

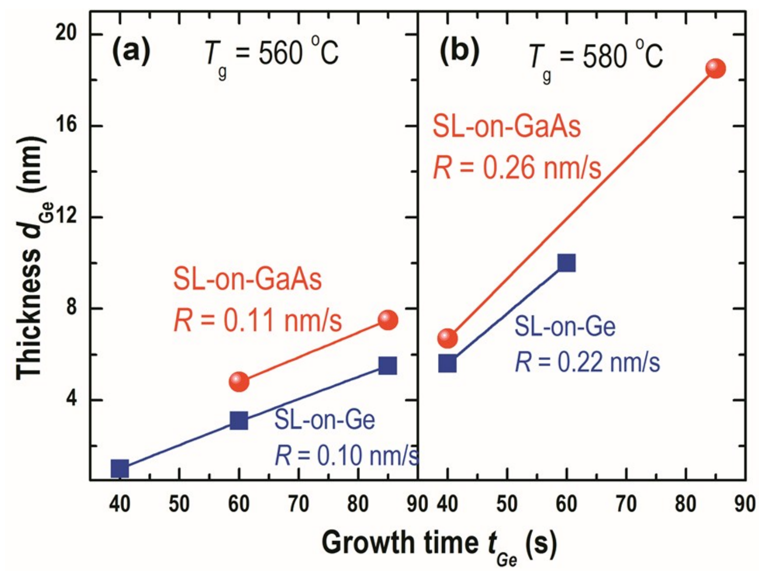

Figure S2 Thickness of the Ge sublayers in the 20-period GaAs/Ge superlattice as a function of growth time with the GaAs growth time fixed at 45 s. The thicknesses were derived from the HRXRD measurements via dynamic theory calculations. The incubation time derived from the plots are in the range of 10~30 s, which agrees reasonable with that reported by Jia et al.,[1] when the growth temperature is taken in to account. Although the data points are limited at certain growth times the resulted thicknesses are repeatable (more than 10 runs were tested).

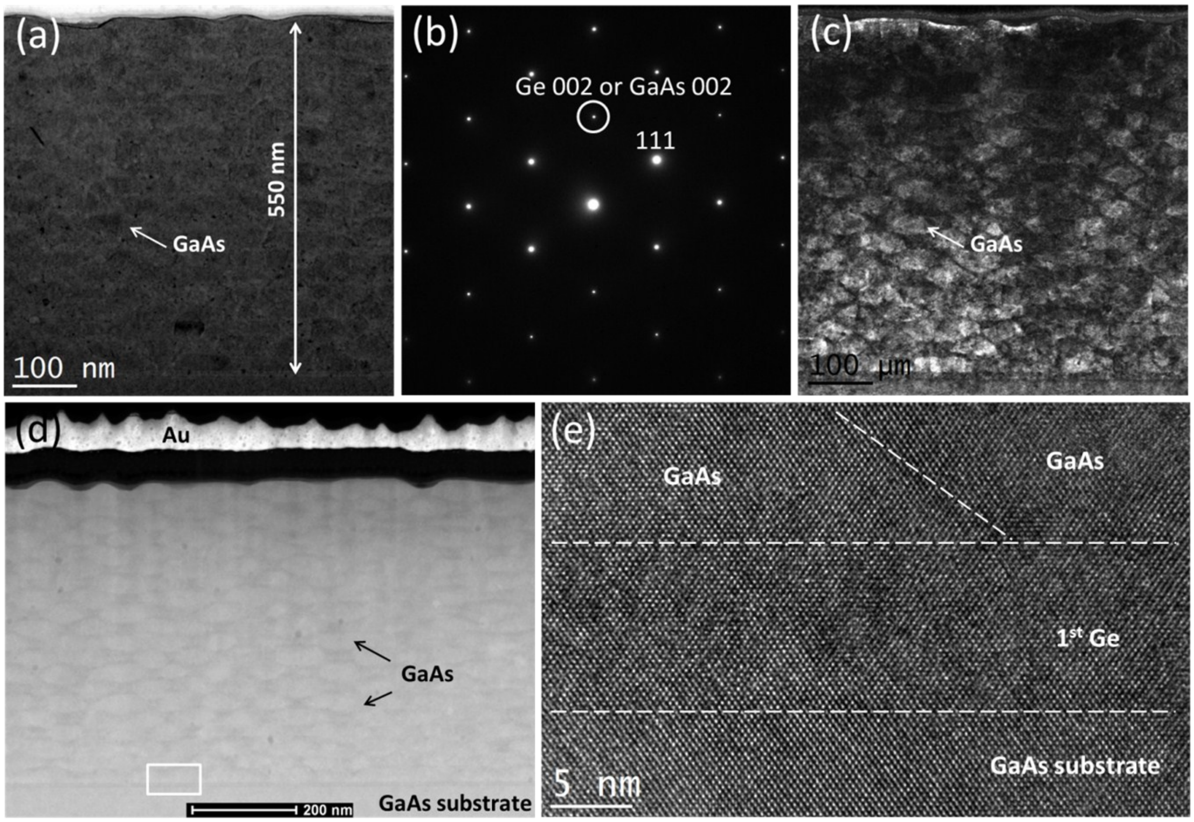

Figure S3 TEM characterization of GaAs/Ge pseudo-superlattice structures grown on GaAs (001) substrate. (a) bright-field TEM image showing the cross-section of the as-grown films, with a thickness measured around ~550 nm; (b) selected-area diffraction pattern taken from the interface area along [1-10] zone axis; (c) dark-field TEM image showing the phase separation with Ge in dark and GaAs in bright; (d) HAADF-STEM image showing the multilayer structures with in-plain undulated 3D island-in-network nanostructures (Ge in dark and GaAs in bright); and (e) high-resolution TEM image showing an epitaxial growth of Ge on GaAs(001) substrate.

Due to the small lattice mismatch between GaAs and Ge (~0.083%), epitaxial growths of Ge-on-GaAs and GaAs-on-Ge with a coherent interface are resulted. This has been confirmed by the diffraction patterns and the HRTEM image shown above in Figure S3b and S3e. We could not observe any superlattice spots between 000 and 002, indicating that the alternative GaAs and Ge layers do not have repeatable thickness in the [002]-direction. Differentiating GaAs and Ge in the bright-field TEM image (i.e., Figure S3a shown above) is quite difficult due to their same crystal structures and close atomic numbers. However, the (002) reflection for fcc Ge is absent while fcc GaAs has a weak (002) reflection owing to a slight difference in the atomic scattering amplitudes between Ga and As. Considering the same contribution from the dynamic diffraction from Ge and GaAs at the zone axes, the Ge layer will appear dark and GaAs is bright in the dark-field TEM images (i.e., Figures S3c and S3d) because more electrons are diffracted in the GaAs crystal. It is interesting to note from dark-field images that the first Ge layer is continuous and uniform in thickness for its nonpolar property, while polar GaAs grows into 3D islands, uniformly in-plane distributed. Thus, the 3D island-in-network nanostructures or a pseudo-superlattice is formed. The pseudo-superlattice has also been demonstrated by the EDX mapping, as shown in the main body of the manuscript.

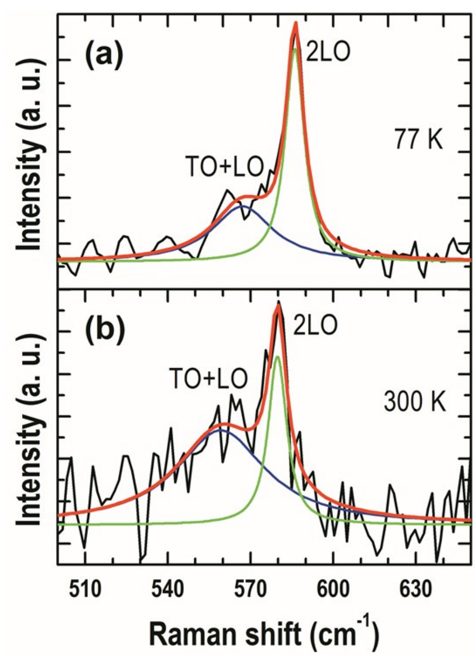

Figure S4 Spectral deconvolution of the representative second-order Raman spectra collected at (a) 77 K and (b) 300 K. They revealed that both TO_{GaAs}+LO_{GaAs} and $2LO_{GaAs}$ overtones have been enhanced as a result of outgoing resonance.

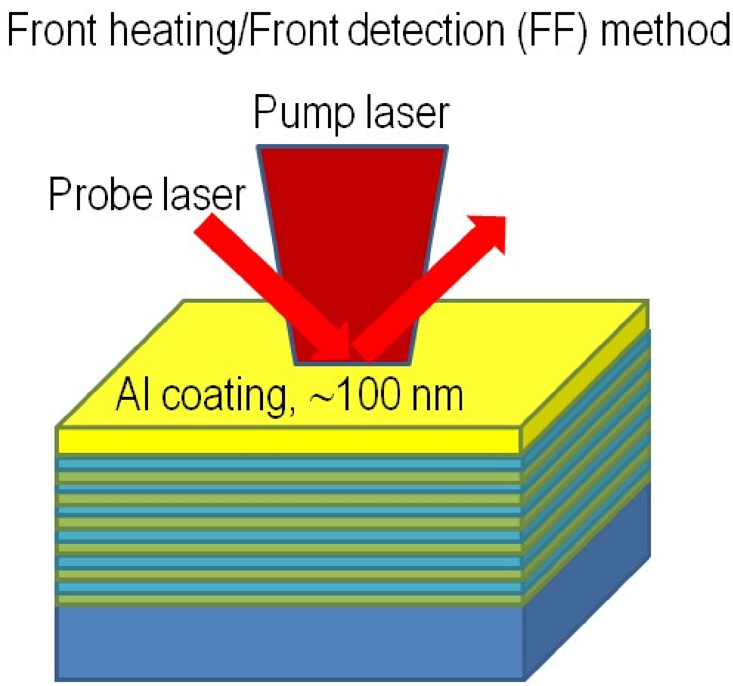

Figure S5 Method employed for measuring the vertical thermal conductivity of the GaAs/Ge superlattice structures as well as the Ge and GaAs wafers that were used as the substrates.

Reference:

1. Jia, R.; Fitzgerald, E. A., *J. Cryst. Growth*, **2016**, *435*, 50-55.