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Supplementary Information for

Droplets induced dot, dot-in-hole, and hole structures in GaGe thin films grown by MOCVD on GaAs substrates

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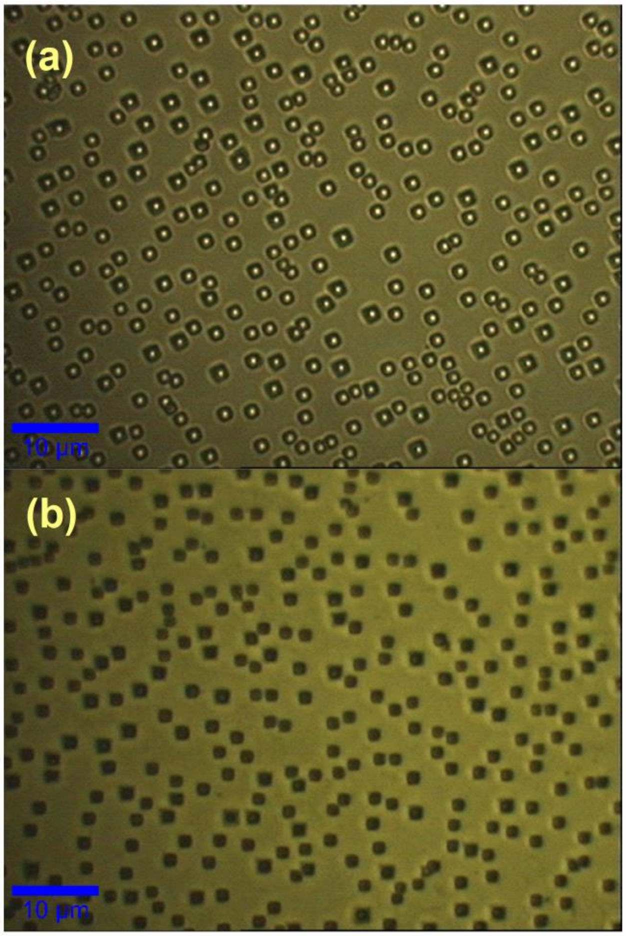

Figure S1 Typical photographs recorded from a GaGe thin film grown at 600 °C before (a) and after (b) ultrasonication process in hot deionized water for 30 min. The scale bars are of 10 μm. Please not the removal of the Ga-rich dots from the dot-in-hole structures after the ultrasonication process.

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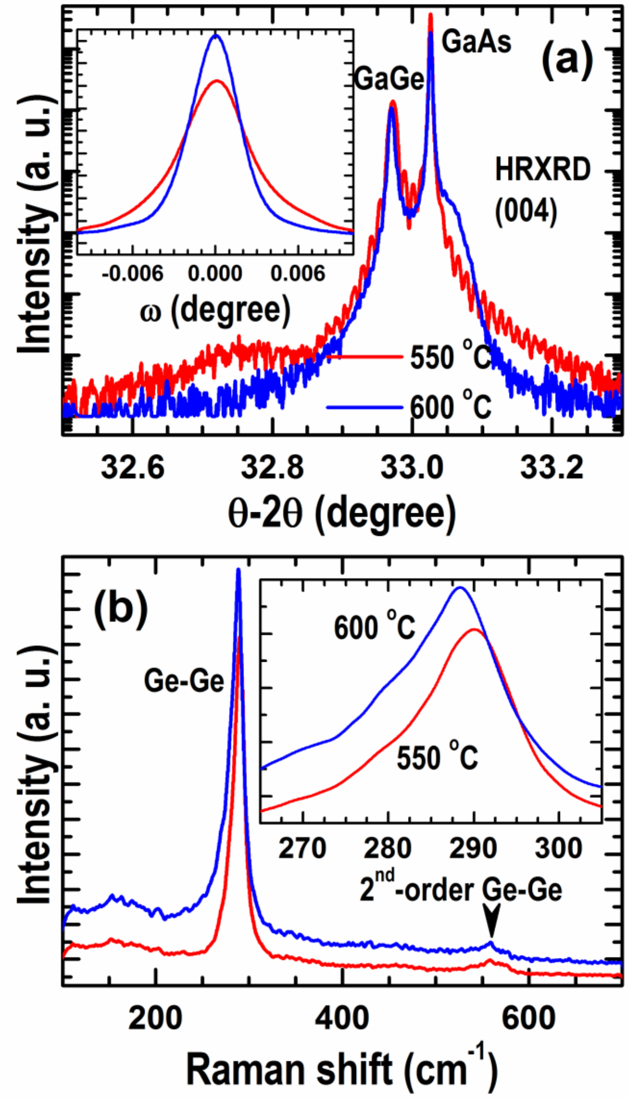

Figure S2 HRXRD (004) diffraction curves (a) and Raman scattering spectra (b) collected from the GaGe thin films grown by MOCVD on GaAs (100) substrates at 550 and 600 °C. The inset in (a) plots the rocking curve of Ga (004) while that in F(b) highlights the Ge-Ge lattice vibrational mode aboyt about 291 cm^{-1}.

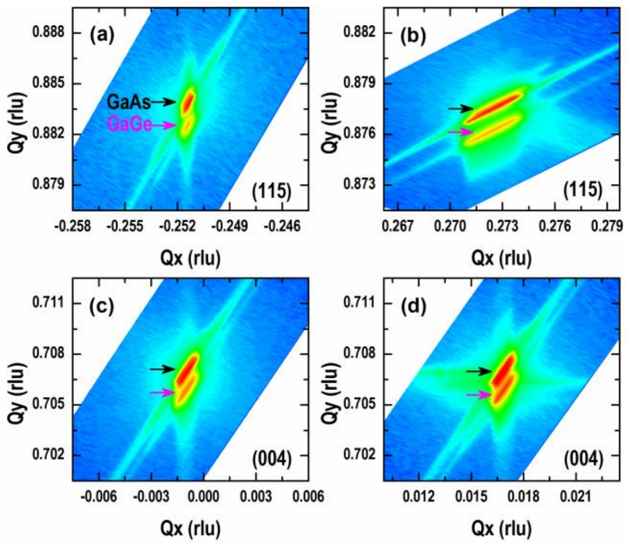

Figure S3 HRXRD reciprocal space mapping from the GaGe thin film samples grown at 550 °C (a) and (c) and 600 °C (c) and (d). (a) and (b) were mapped aiming at the (115) planes and (c) and (d) were mapped aiming at the (004) planes. Please note that (a) was mapped with high incident angle while (b) was with low incident angle. The alignment between the GaAs (115) and GaGe (115) in Qx indicates the coherent compressive strain of the GaGe layer epitaxially grown on the GaAs substrates.

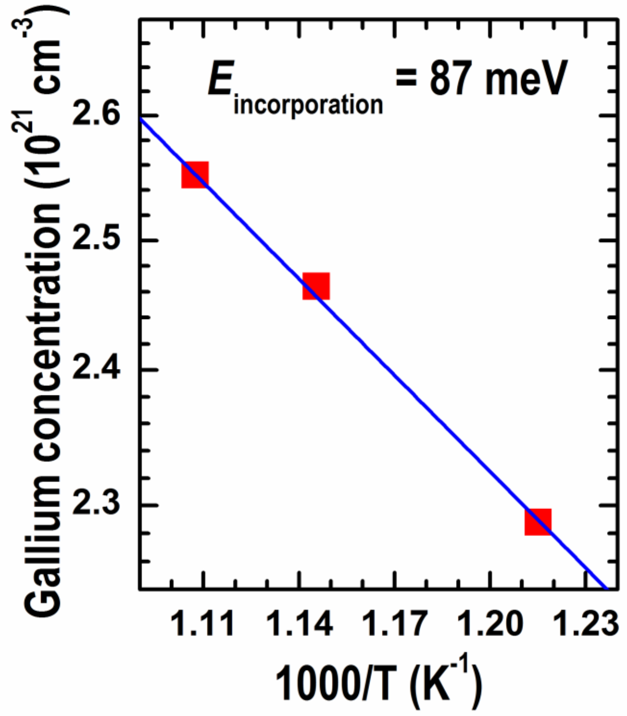

Figure S4 Arrhenius plot of gallium concentrations in the GaGe thin films probed by point-focus EDX from the flat areas away from the droplet-induced features as a function of growth temperatures. An activation energy of 87 meV is obtained from the Arrhenius plot, which much smaller than the surface migration characteristic energy (2.85 eV) of the Ga-rich droplets [see Fig. 1(d) and the context for details], indicating that the incorporation of Ga atoms into the GaGe thin film is directly from the Ga-rich liquid rather than via surface diffusions.

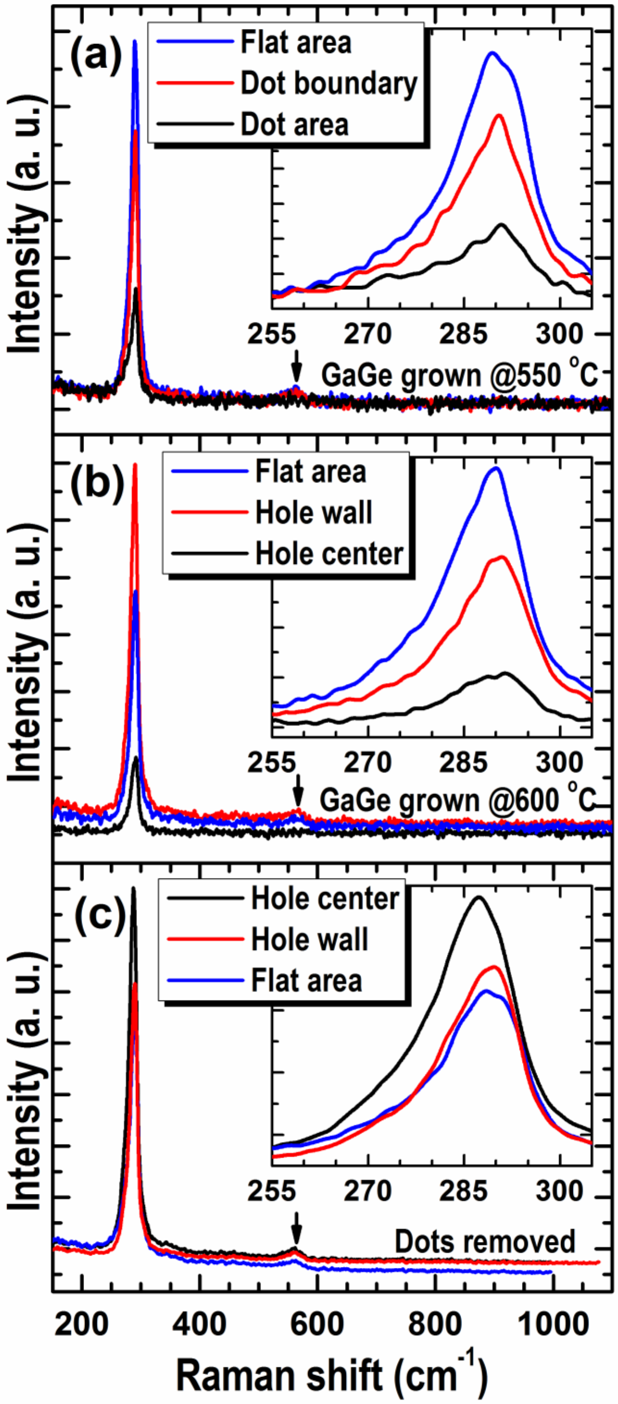

Figure S5 Raman spectra collected from the GaGe thin films samples grown at 550 °C (a), 600 °C (b), and 600 °C followed by an additional ultrasonication process in hot (80 °C) deionized water (c). The insets highlight the Ge-Ge lattice vibration mode addressing their intensity evolutions across the dot, dot-in-hole, and empty hole structures. The arrows indicate the second-order mode of the Ge-Ge vibrations.

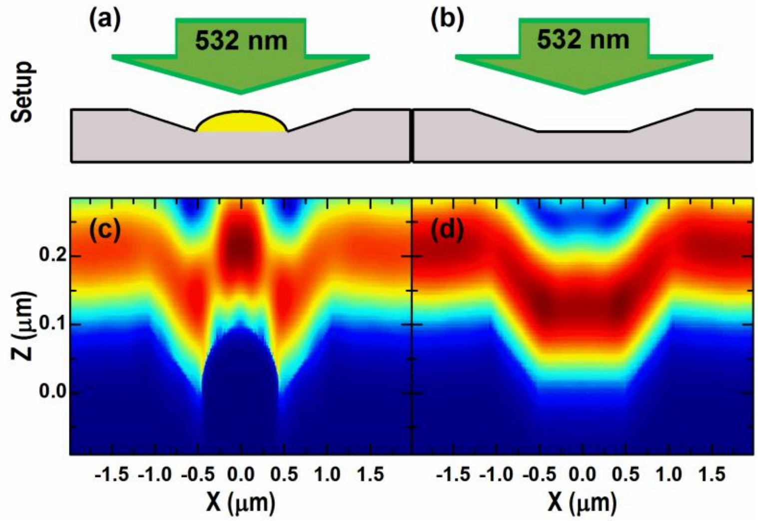

Figure S6 (a) and (b) schematic diagrams showing the configuration and setup employed in the finite-difference time-domain (FDTD) simulations for germanium holes with and without the gallium dot; (c) and (d) distributions of electrical field magnitudes derived from the FDTD simulations. The magnitude evolutions along the vertical direction, *i.e.*, Z-axis, provide evidence for the interference between the incident and reflected waves. The constructive interference gives rise to the enhanced electrical field in the germanium holes.