

**Fig S1**: Schematic of droplet diffusion and crystallization processes during group-V exposure of the group-III droplet.

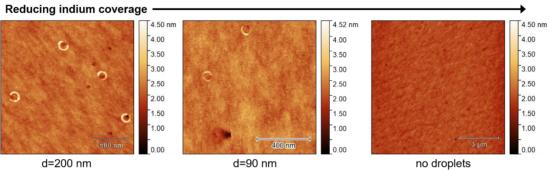


Fig S2: AFM images of samples with reduced group-III deposition. Droplet diameter decreases with group-III coverage until no droplets are present. All samples exhibit ring-like structures.

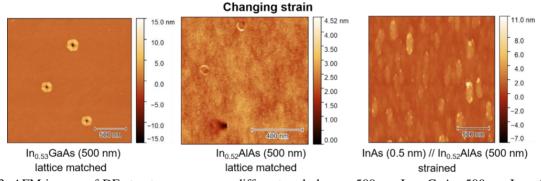


Fig S3: AFM images of DE structures grown on different underlayers:  $500 \text{ nm } In_{0.53}GaAs$ ,  $500 \text{ nm } In_{0.52}AlAs$ , and  $0.5 \text{nm } InAs/500 \text{ nm } In_{0.52}AlAs$ .

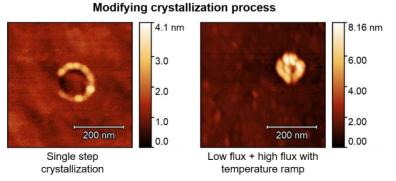


Fig S4: AFM images of DE structures grown on with different crystallization steps. A single-step crystallization process takes place with maximum As flux and at the same temperature as the droplet deposition. A two-step crystallization exposes the droplet to a low As flux, followed by the maximum As flux with a temperature ramp.

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