

Digital Transfer Growth of Patterned 2D Metal Chalcogenides by Confined Nanoparticle Evaporation

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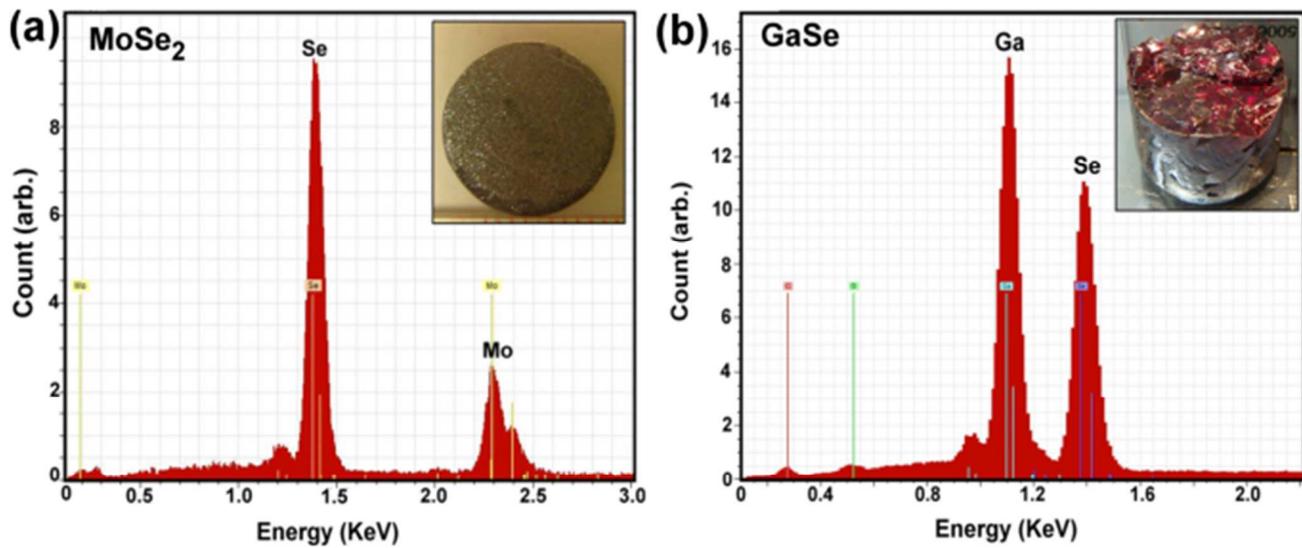


Figure S1. EDX spectra and photographs of bulk targets used for laser ablation. (a) MoSe₂ target with a 1:2 elemental ratio of Mo and Se. (b) GaSe target with a 1:1 elemental ratio of Ga and Se.

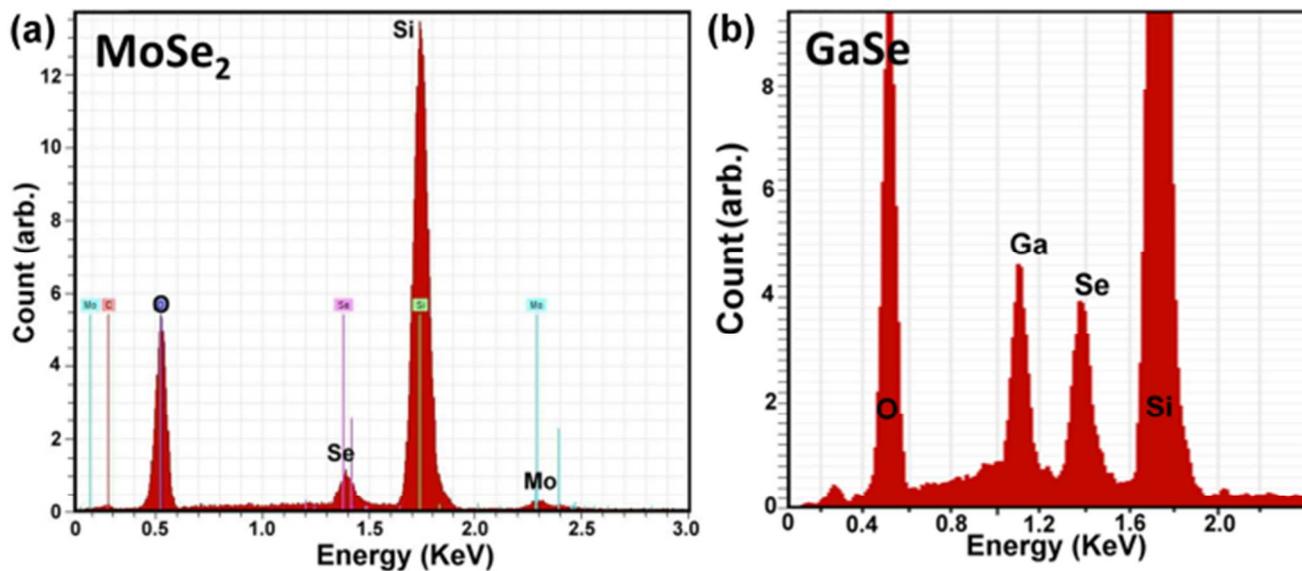


Figure S2. EDX spectra of PLD-deposited nanoparticles on SiO₂ (300 nm)-coated Si substrates. (a) MoSe₂ nanoparticles, maintaining a 1:2 elemental ratio of Mo and Se (b) GaSe nanoparticles, maintaining a 1:1 elemental ratio of Ga and Se within instrumental errors of $\pm 3\%$.

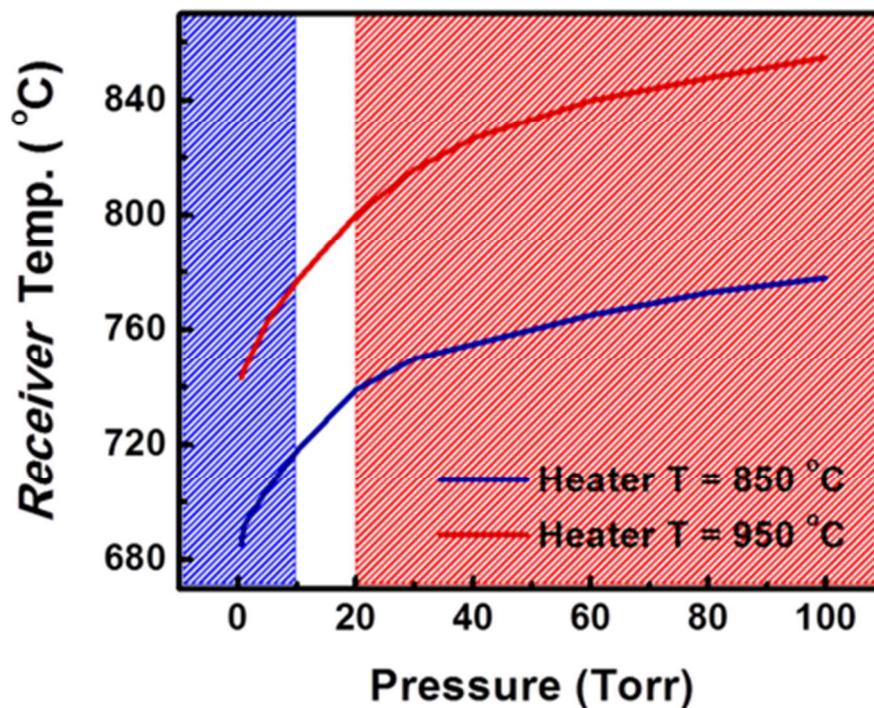


Figure S3. Temperature of the receiver substrate used for 2D crystal growth which was tuned, after setting the heater to a temperature that would evaporate the precursor (~ 850 °C for GaSe and ~ 950 °C for MoSe₂), using Ar pressure within the chamber to adjust the thermal coupling between the source and receiver substrates. The temperatures of the source and receiver substrates were measured by a thermocouple and a pyrometer, respectively. A pressure of 10-20 Torr was required to achieve a growth temperature of ~ 730 °C for GaSe and ~ 800 °C for MoSe₂. Pressures below 10 Torr in both cases resulted in a receiver substrate temperature that was too cool for 2D crystal growth, yielding amorphous films. Conversely, for pressures above 20 Torr, the receiver substrate was too hot for growth, and material simply evaporated from the receiver.

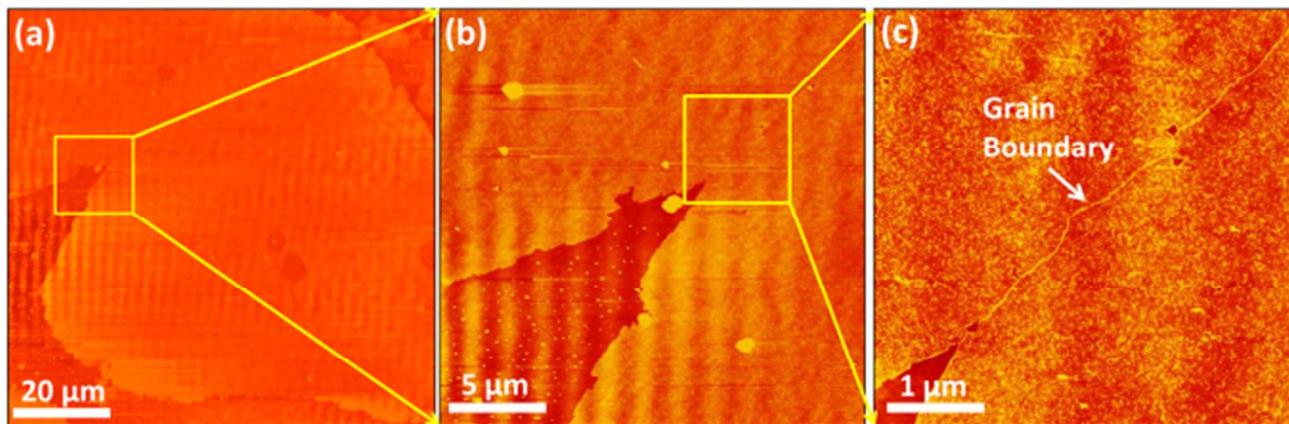


Figure S4. AFM images of GaSe grown by digital transfer growth showing the grain boundaries observed between two large monolayer nanosheets after they coalesce. Higher magnification scans (as in (c)) revealed observable grain boundaries. Line scans of the grain boundary showed topographical discontinuities less than 1 nm in height.

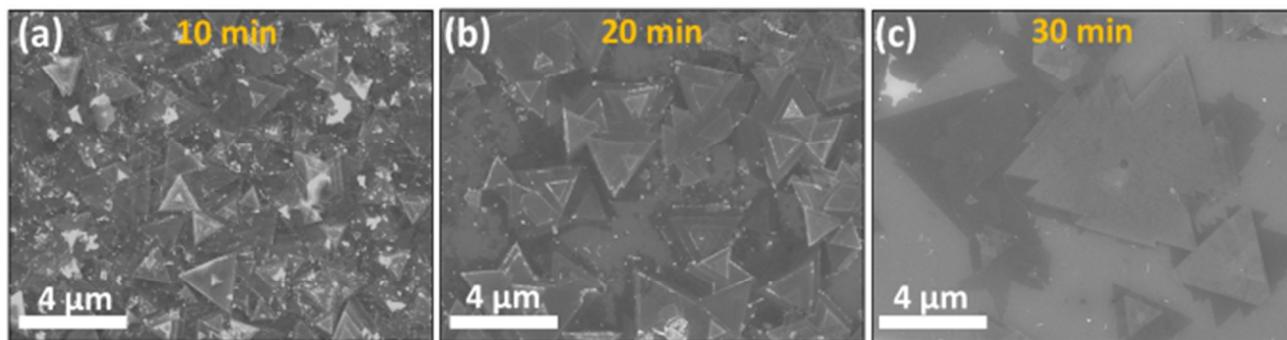


Figure S5. SEM images showing the crystallite sizes measured on the receiver substrate resulting from varying digital transfer growth times of 10-30 min at a heater temperature of 850 °C and Ar pressure of 20 Torr for identical GaSe nanoparticle depositions of 50 laser pulses. (a) High density nanosheets with lateral sizes of $\sim 2 \mu\text{m}$ after 10 min growth time. Growth times of (b) 20 minutes and (c) 30 minutes resulted in the formation of larger flakes with lateral sizes of 4-10 μm , respectively.