## Enhancement of superconductivity in cryogenically grown ultra thin Al films

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Superconductivity in thin films can deviate significantly from bulk behavior, especially as dimensionality and disorder come into play. This is particularly true for aluminum, where critical temperature (T<sub>C</sub>) and film morphology are highly sensitive to thickness and growth conditions. Here, we present an *in-situ* scanning tunneling microscopy (STM) study, performed at 78 K, of Al thin films grown on atomically clean Si(111) substrates by molecular beam epitaxy at cryogenic temperatures down to 6 K. The morphology is characterized across a wide range of coverages, from sub-monolayer up to 20 monolayers (ML). Cryogenic growth results in oriented hexagonal islands that begin to coalesce into a continuous film around 5 ML, with a typical roughness of a few monolayers. This roughness is constant up to 20 ML. Upon annealing to room temperature, the surface becomes nearly atomically smooth, though grain boundaries remain visible in STM. In contrast, room temperature growth produces significantly rougher films with large, disconnected islands of varying shape and orientation. We also investigated the superconducting properties of cryogenically grown films after exposure to atmospheric conditions, as required for ex-situ transport measurements. To stabilize the films, we used different post-growth treatments, including low temperature capping, cold oxidation, and room temperature oxidation. The films show critical temperatures approaching 3 K and critical fields above 5 T, which are significantly above the bulk value of 1.2 K.

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## **Supplementary information:**

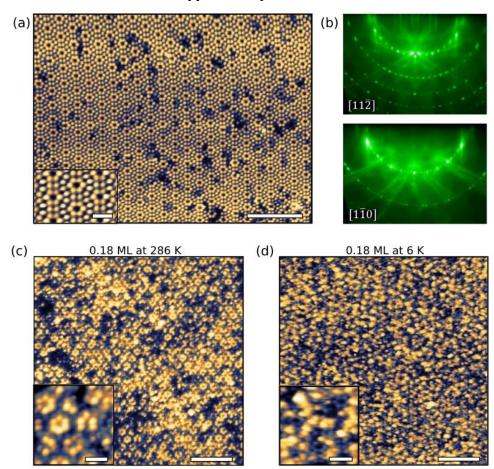


Figure 1. (a) STM image of a clean Si(111) surface with 7 x 7 surface reconstruction also confirmed by (b) RHEED. Initial stages of the growth at 0.18 ML of Al compared at growth temperatures of (c) 286 K and (d) 6 K.

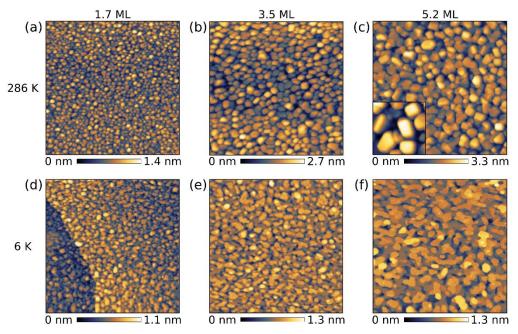


Figure 2. Evolution of the growth at 286 K for (a) 1.7 ML, (b) 3.5 ML, and (c) 5.2 ML. (d)-(f) This is compared to growth at 6 K for the same thicknesses