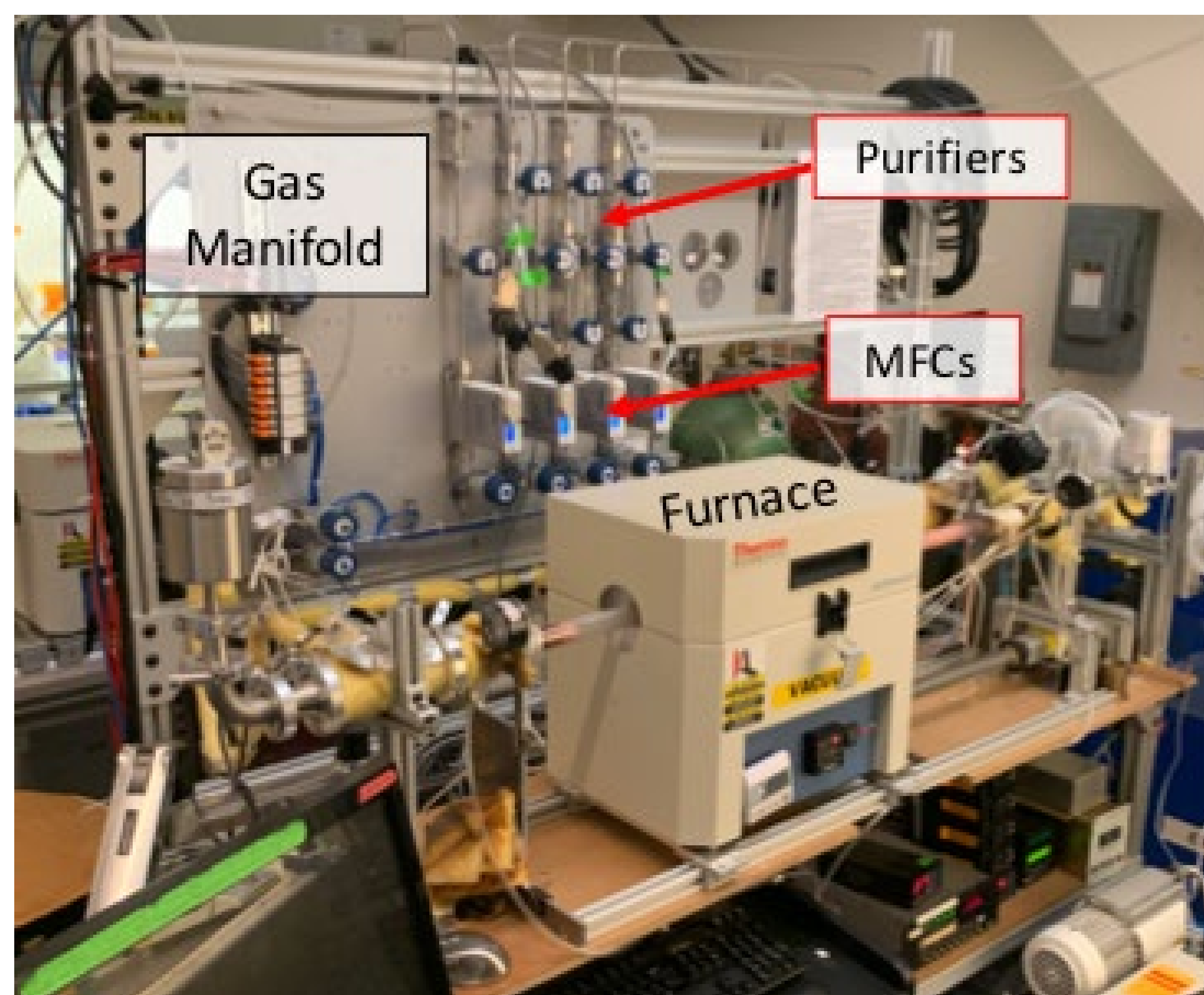


# Epitaxial ultraclean, wrinkle-free graphene growth on Cu(111) in an oxygen-free environment

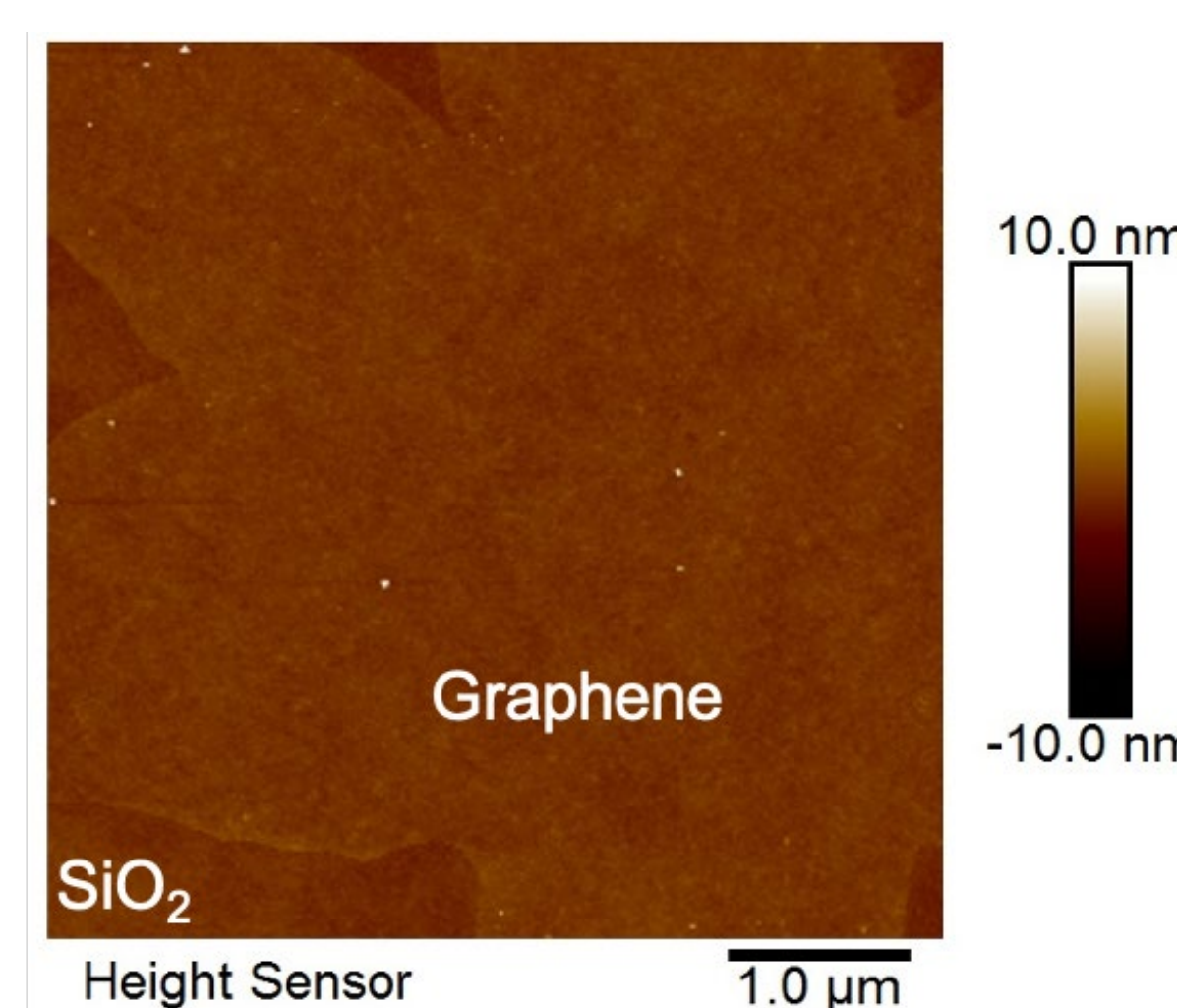
Chemical vapor deposition (CVD)-derived graphene performance has shown to deteriorate with surface wrinkles, folds, and transfer-related contaminations. Towards the stitching-up phase of the graphene growth, the lack of active catalytic copper surface slows down the growth rate and leads to an excess of amorphous carbon formation. With the integration of an oxygen-free growth environment and a Cu(111) growth substrate; flat, clean, and intrinsic defect-free graphene can be reproducibly grown with an enhanced growth rate via low pressure CVD (LPCVD). The resulting sheet of graphene shows an epitaxial relationship with the substrate. Contamination-free graphene surface also enables clean transfer due to the absence of amorphous carbon and structural defects in the graphene sheet. Electrical measurements with h-BN encapsulation demonstrates carrier mobility comparable to exfoliated graphene, with ballistic transport characteristics at low temperature.

## Oxygen Free System Design and Growth Quality

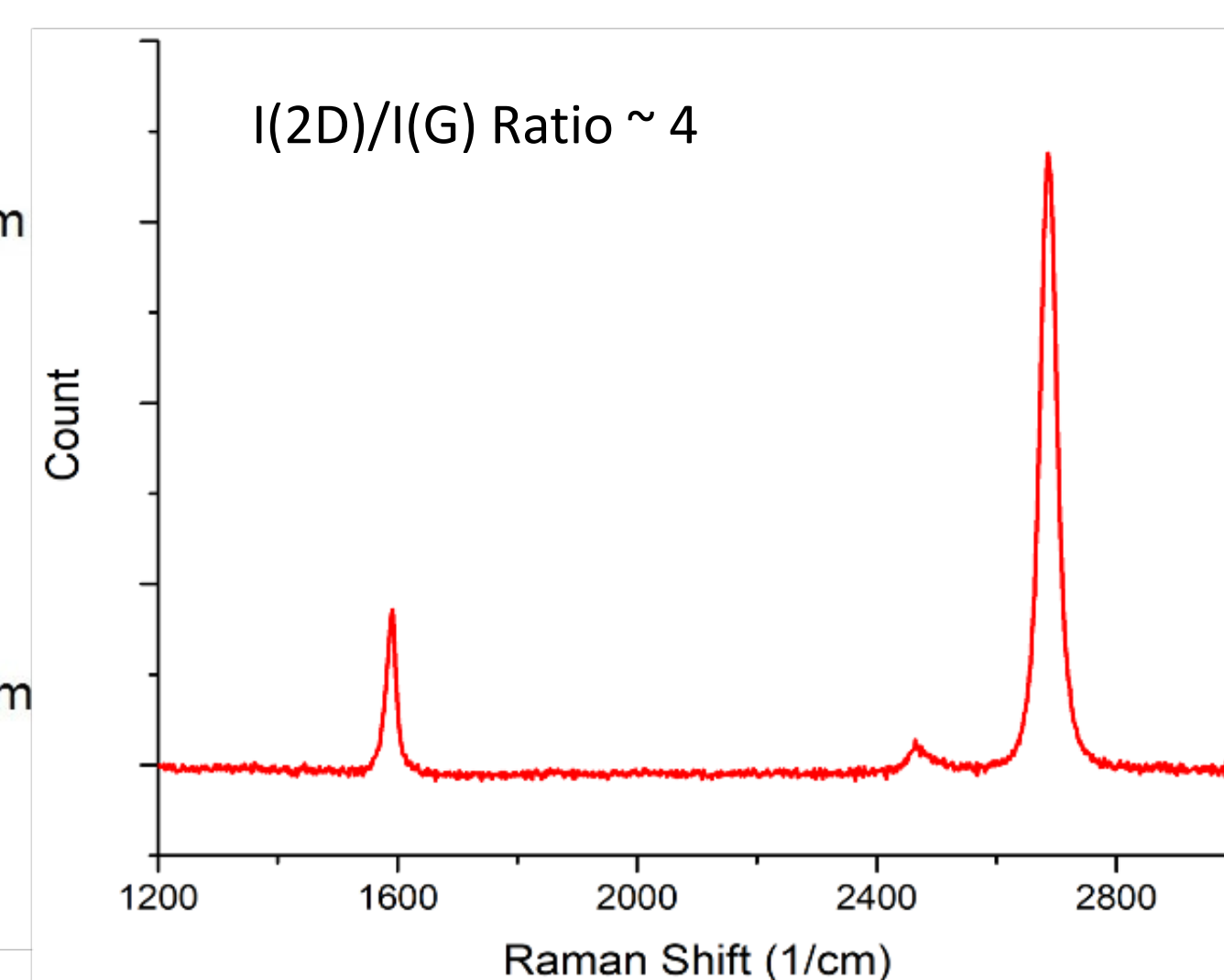


System Layout

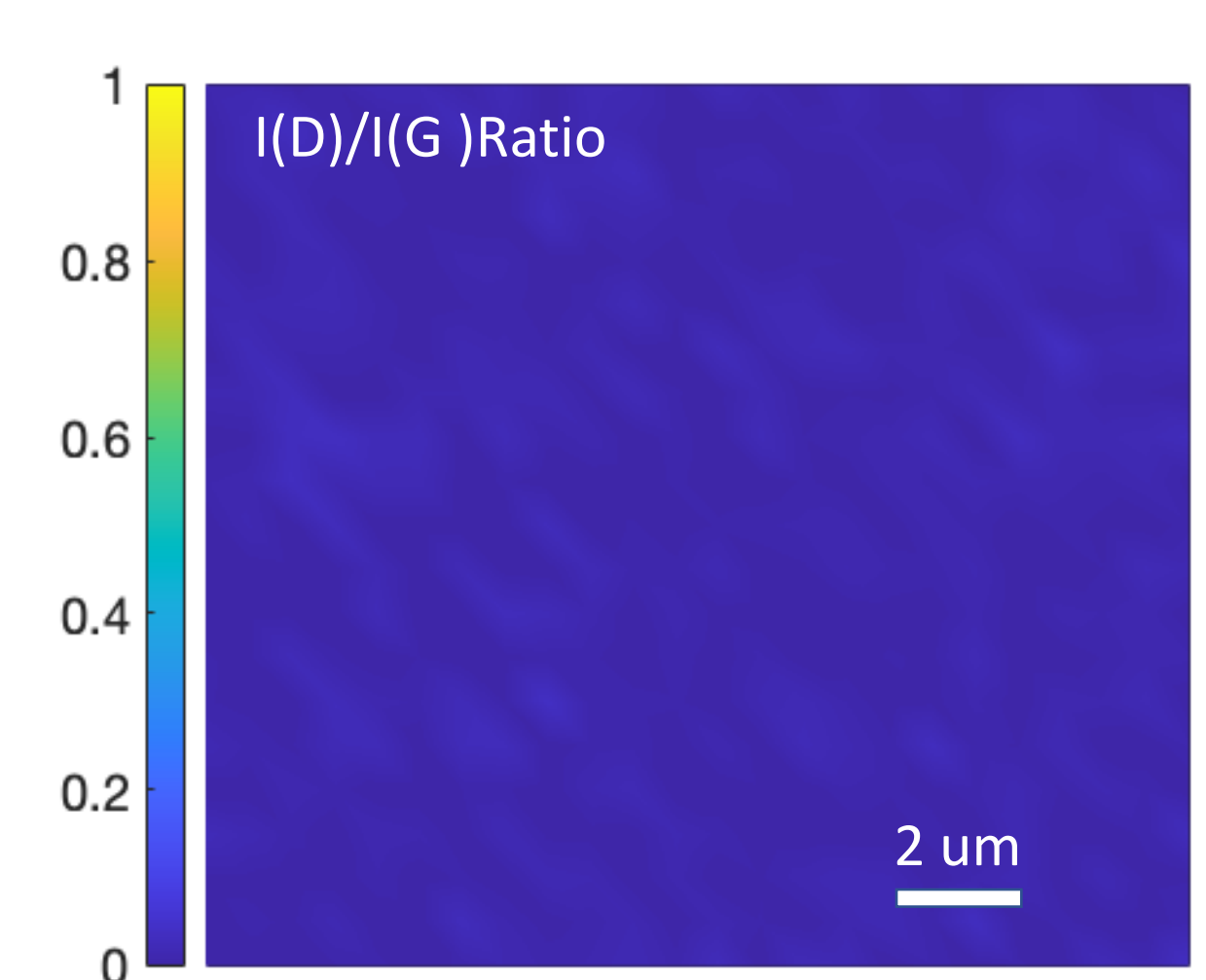
- ULSI grade gas feedstock (99.9999% purity)
- Gas purifiers to reduce oxygen species from gas source ( $O_2 < 1$ ppb)
- Helium-leak detection to optimize system leak integrity
- Load-lock installed to minimize pump down and  $O_2$  contamination



Residue free transfer

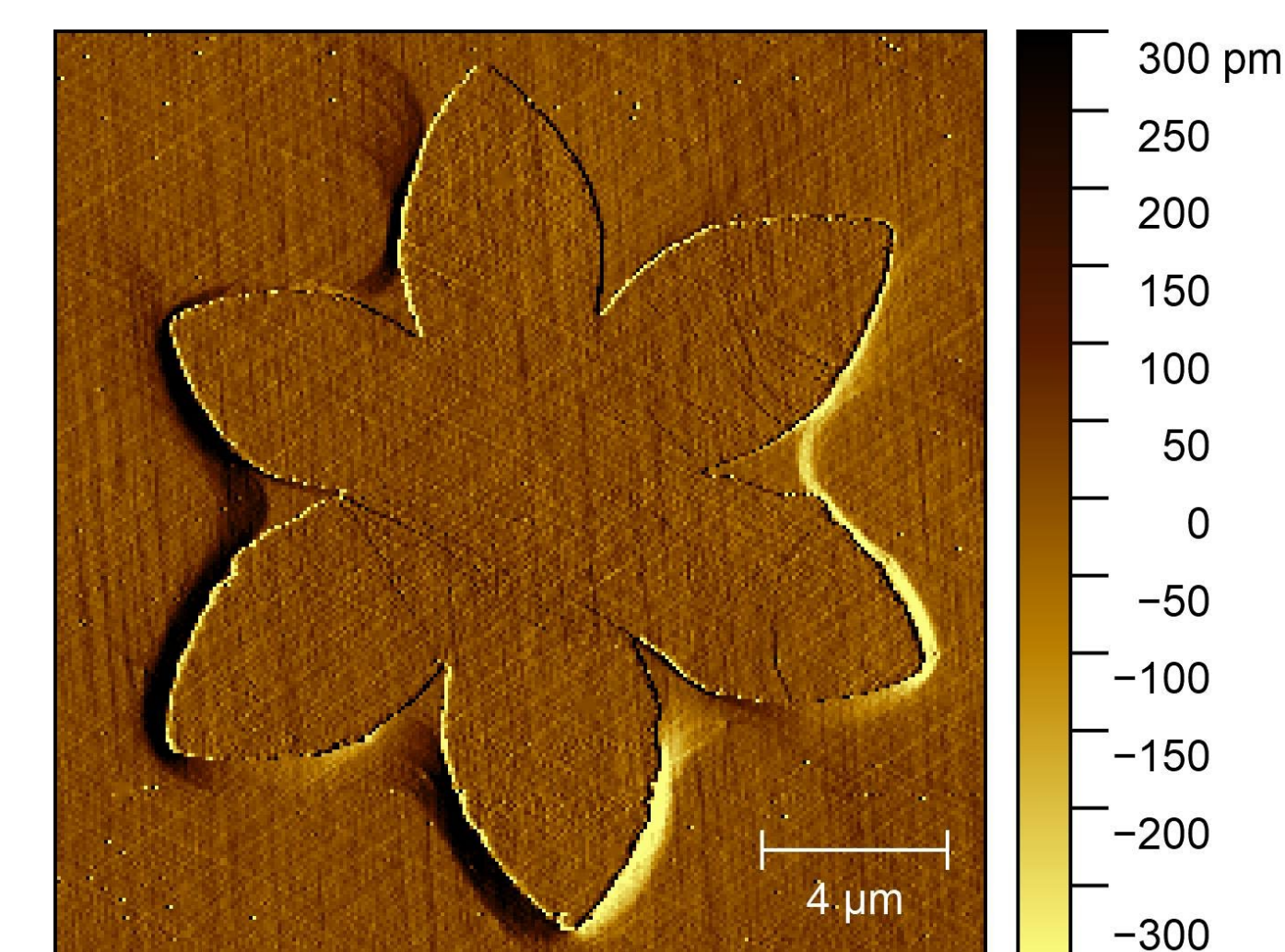


Raman Characterization

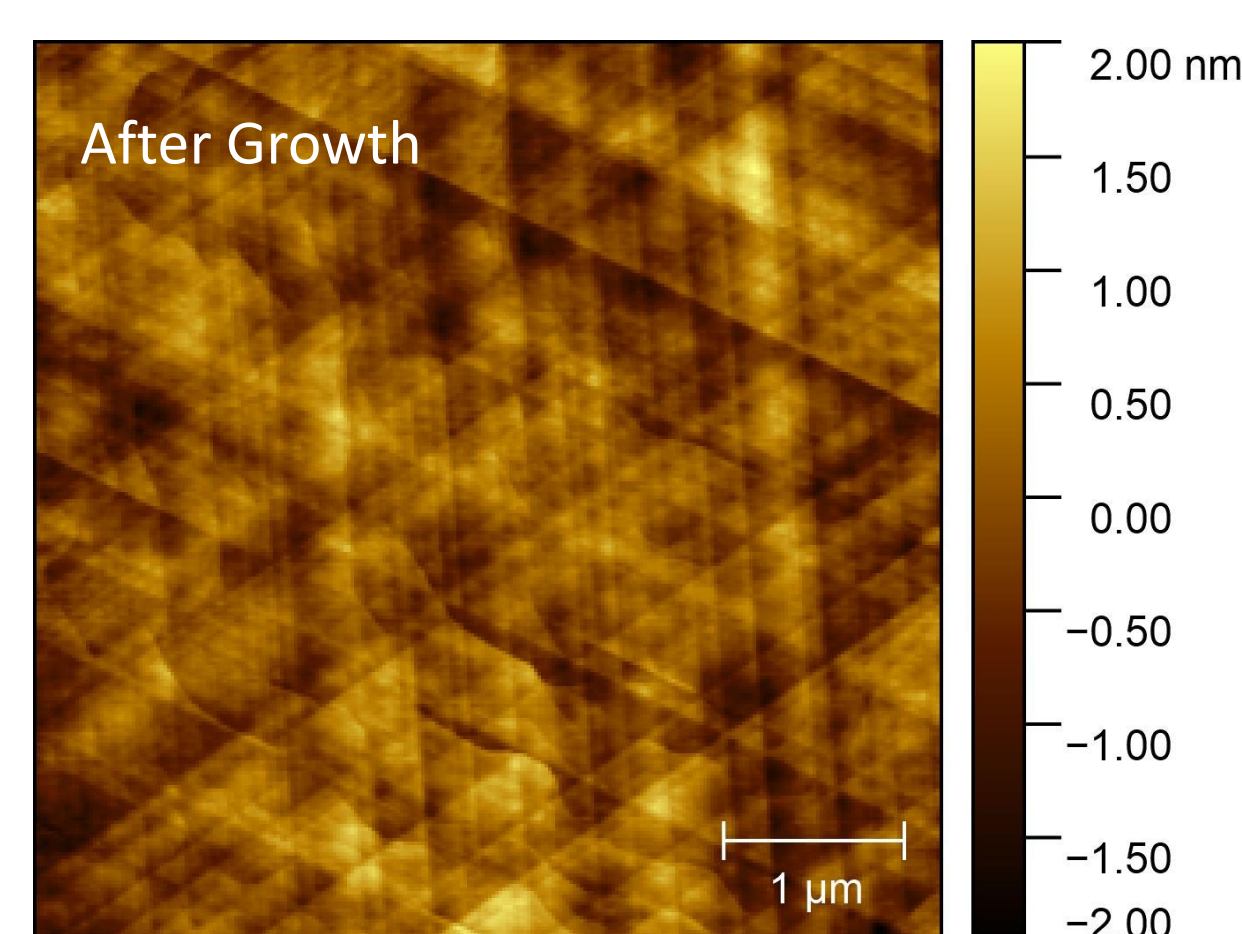
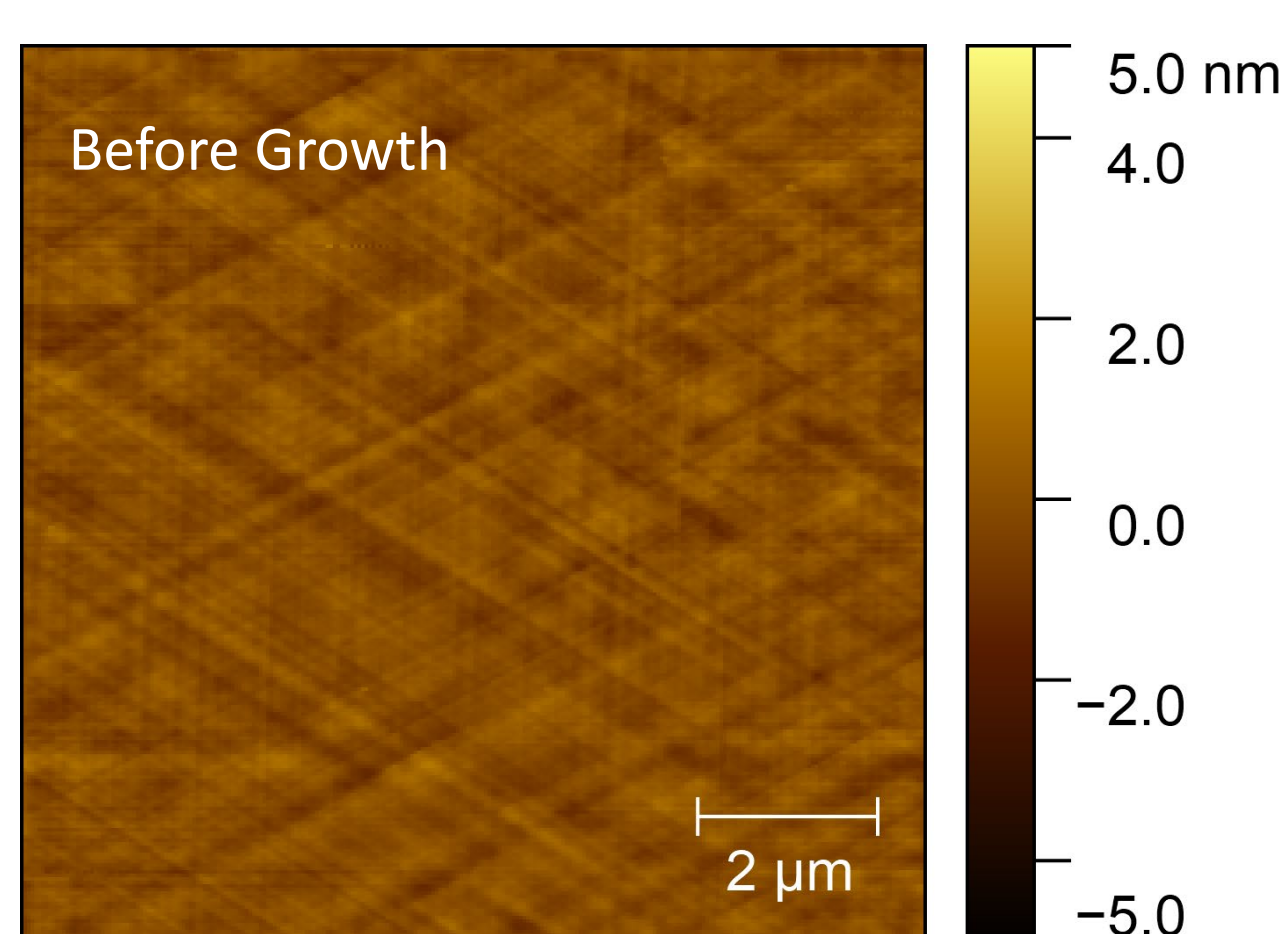


## Single crystal Cu(111) substrate for high quality epitaxial graphene growth

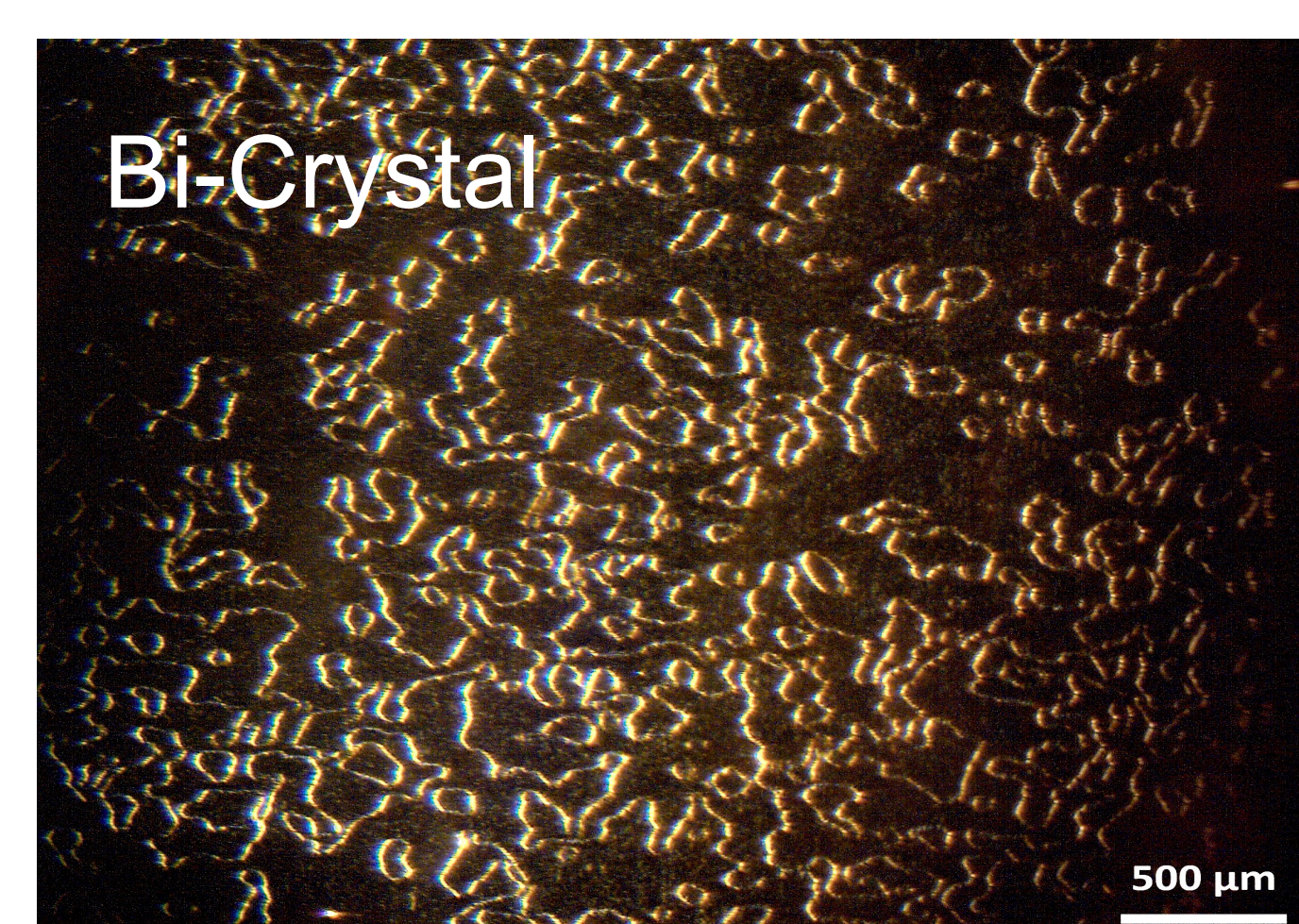
- 12h anneal in atmospheric pressure oxygen environment before sputtering copper
- DC sputtering copper ~ 1000 nm
- Post annealing in  $H_2$  / Ar environment for 4h to recrystallize film to single crystal
- Preserved surface flatness after growth (~ 2nm roughness)
- $O_2$  annealing produces better crystallinity



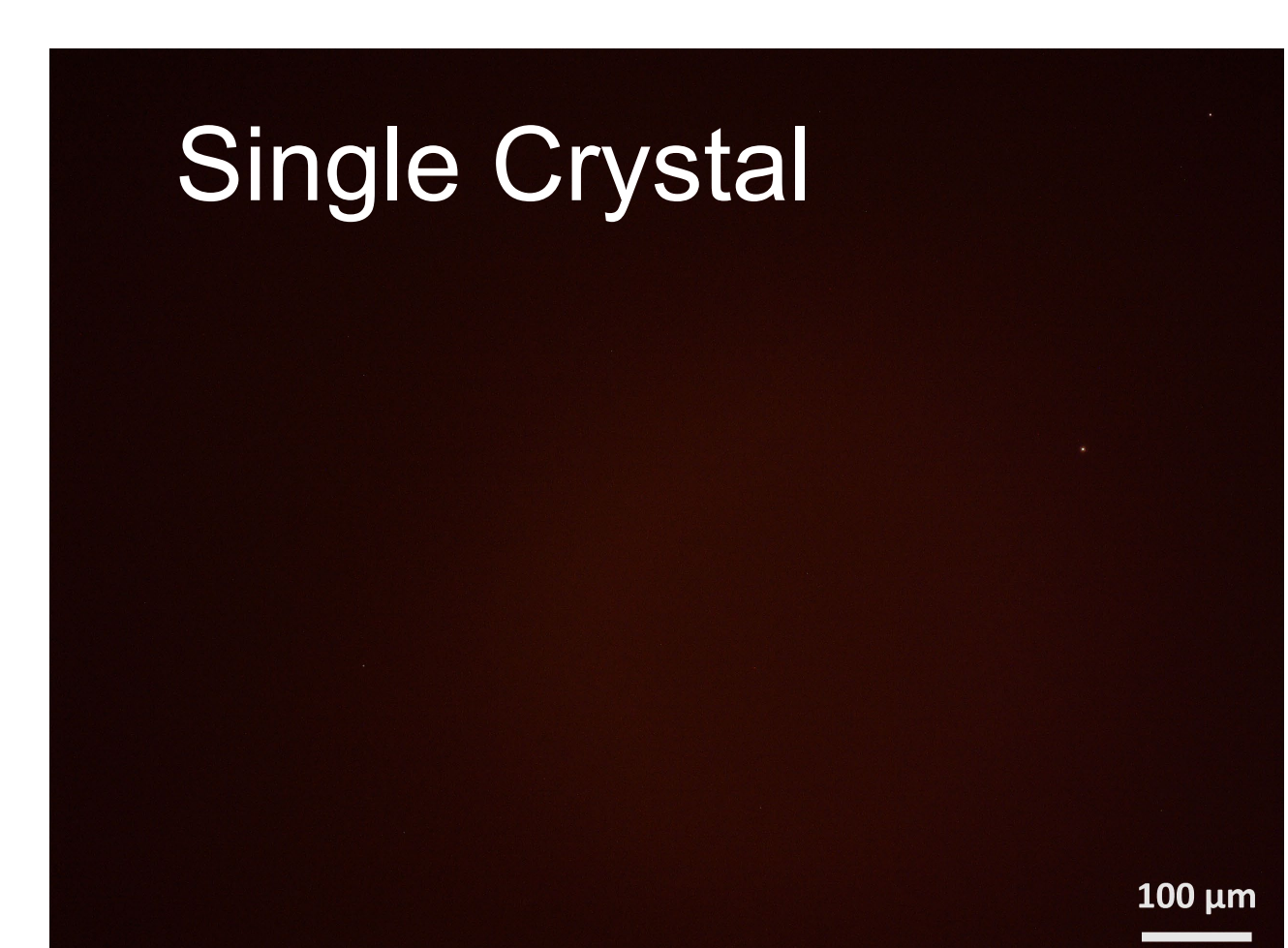
Aligned grains on substrate



Surface roughness is preserved after growth



Acid Pretreatment



Oxygen Pretreatment

### Contact Info

Xingzhou Yan  
[xy2376@columbia.edu](mailto:xy2376@columbia.edu)  
 Jacob Amontree  
[jma2260@columbia.edu](mailto:jma2260@columbia.edu)

1. Lin, L. et al. Nat. Commun. 10, 1912 (2019).
2. Deng, B. et al. ACS Nano 11, 12337–12345 (2017).
3. Wang, M. et al. Nat. 2021 5967873 596, 519–524 (2021).
4. Zhang, J. et al. ACS Nano 14, 10796–10803 (2020).
5. Burton, O. J. et al. ACS Nano 14, 13593–13601 (2020).