# **Atomic Scale Investigation of CVD Graphene Growth Mechanism on Cu(111)**

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### **Evolution of CVD graphene on Cu**



Figure 1. Top: Schematic illustration of surface intermediates at different reaction conditions. Bottom: Overview of STM images showing (A) Carbon clusters; (B) the boundary between carbon clusters and defective graphene; (C) defective graphene; (D) vacancy-free graphene.





Figure 2. (a) A Cu terrace fully decorated by carbon clusters; (b)~(f) **Enlarged STM images showing different types of carbon clusters:** dimer, rectangle ; zigzag and armchair-like chain. (g) Relative ratio of the carbon clusters; (h) Ordered alignment of carbon clusters with respect to substrate lattice; (i)optimized structure of carbon rectangle on Cu(111) and (j) the simulated STM image

Figure 3. (a) STM topography image of ~0.8ML of defective graphene and (b) its corresponding conductance image; (c) domain boundary between the carbon clusters and defective graphene; (d) Defective graphene on two terraces; (e) defective graphene with pseudo-periodic corrugations; (f) typical vacancies at the intersection of the corrugations.

## Conclusion

Intermediates of CVD graphene on Cu: dimer, carbon rectangle; zigzag and armchair-like carbon chain;

**D** Saturation of carbon clusters give rise to defective graphene: pseudo-ordered corrugations and vacancies;

**Only annealing under methane can heal the defects.** 

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## **Defects Healing**



#### Figure 4. (a) Vacancy-free graphene after healing at 800° C in the presence of methane; (b) moiré periodicity; (c) atomic resolution