Layer and Stacking Controlled Epitaxial Growth of Graphene Films

Luzhao Sun[1]*, Buhang Chen[1], Yanyan Dong[1], Chengjin Wu[1], Zhongfan Liu[1,2]
[1]*Beijing Graphene Institute, No.13 Cuihu Nanhuan Road, Sujiatuo Town, Haidian District, Beijing, China*
[2]*College of Chemistry and Molecular Engineering, Peking University, Chengfu Road 202, Haidian District, Beijing, China*

sunlz@bgi-graphene.com

Abstract

Compared to their monolayer counterpart, bilayer, trilayer, and few-layer graphene films offer greater application potential because of their enhanced band structure tunability, improved electrical and thermal properties, and superior mechanical strength and barrier performance. However, the layer- and stacking order-controlled synthesis of graphene films remains challenging: 1) The dangling-bond-free nature of two-dimensional surfaces impedes the nucleation of additional layers and prevents true layer-by-layer growth, inevitably leading to thickness variations via the Stranski-Krastanov mode; 2) The energetically favorable Bernal stack configuration impedes the formation of twist-stacking order[1]. This talk will present our recent progress in this field. We propose two distinct growth modes: 1) layer-by-layer growth for fractional-layer graphene films[2], and 2) multi-layer synchronous epitaxial growth for growing well-defined integer-layer films[3]. A hetero-site nucleation strategy that triggered by gas-flow perturbation during graphene growth affords twist angles ranging from 0° to 30°[4]. By designing novel heterogeneous catalyst substrates and precisely regulating growth thermodynamics and kinetics, we achieved the precise synthesis of 2-7 layer graphene films, the industrial-scale production of homogeneous A3-sized trilayer graphene films (42 × 30 cm^2)[3], the roll-to-roll growth of meter-sized fractional-layer graphene films (2-5 layers)[2], and 2-inch trilayer graphene single-crystal wafers[5]. Our established methods open new paradigms for the layer-controlled synthesis of 2D materials films and would inspire the atomic-level synthesis and manipulation of nanomaterials.

References

[1] L. Sun, G. Yuan, L. Gao*, J. Yang, M. Chhowalla*, M. H. Gharahcheshmeh, K. K. Gleason*, Y. S. Choi, B. H. Hong*, Z. Liu*, *Nat. Rev. Methods Primers*, 1 (2021) 5.
[2] Y. Dong, B. Chen, C. Wu, Y. Chen, A. Zhu, Q. Li, P. Bian, X. Song, L. Jia, S. Zheng, Q. Cai, Y. Li, X. Song, J. Wang, M. Li*, L. Sun*, Z. Liu*, *Nano Res.* 18 (2025), 94907558.
[3] B. Chen, X. Zeng, Z. Liu, W. Dong, D. Pei, H. Wang, Y. Dong, C. Wu, X. Gao, H. Xiao, H. Gao, H. Jia, A. Yuan, J. Du, H. Chen, H. Liu, C. Tan, J. Yin, Z. Liu, L. Liu, P. Gao, K. Novoselov, H. Peng, Z. Li*, L. Sun*, Z. Liu*, *Nat. Commun.* 16 (2025) 5490.
[4] L. Sun, Z. Wang, Y. Wang, L. Zhao, Y. Li, B. Chen, S. Huang, S. Zhang, W. Wang, D. Pei, H. Fang, S. Zhong, H. Liu, J. Zhang, L. Tong, Y. Chen, Z. Li, M. H. Rümmeli, K. S. Novoselov, H. Peng*, L. Lin*, Z. Liu*, *Nat. Commun.* 12 (2021) 2391.
[5] C. Wu, B. Chen, H. Liu, X. Song, S. Zheng, Q. Li, Y. Dong, S. Li, J. Shao, P. Bian, J. Xue, X. Huang, X. Sun, K. Jia*, W. Wei, Z. Gao, L. Sun*, Z. Liu*, *Adv. Mater.* 37 (2025) 2501582.

Figures

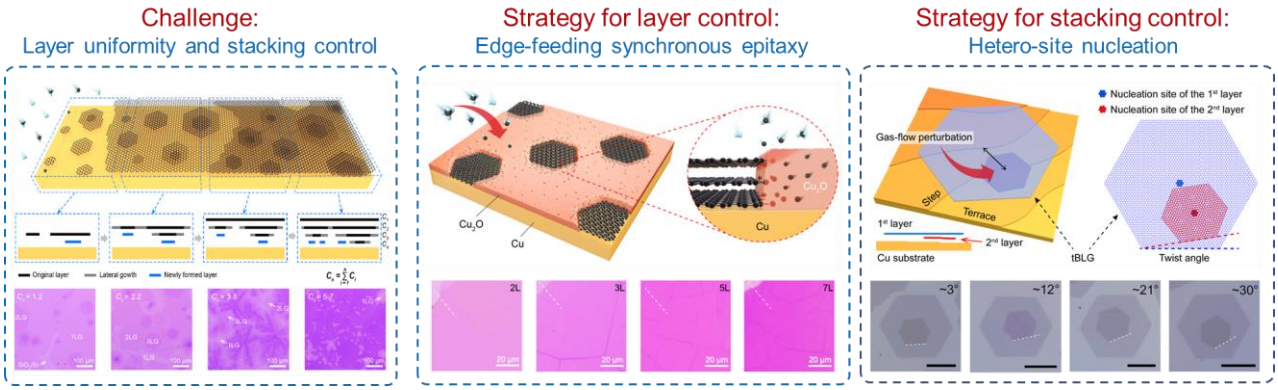

Figure 1: Layer and stacking controlled growth of graphene