Single Crystal, Large-area, Fold-free Monolayer Graphene

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Abstract

Chemical vapor deposition of carbon-containing precursors on metal substrates is currently the most promising route for the scalable synthesis of large-area high-quality graphene films. However, there are usually some imperfections present in the resulting films: grain boundaries, adlayers and wrinkle/folds, all of which can degrade the performance of graphene in applications. Numerous studies have been made on ways to eliminate grain boundaries and adlayers, but graphene folds have been less investigated (and in any case, not heretofore eliminated). Here we explored the wrinkling/folding process for graphene films grown from an ethylene precursor on single-crystal Cu-Ni(111) foils. We identified a critical growth temperature (1030 K) above which folds will naturally form during the subsequent cooling process. (Specifically, the compressive stress that builds up due to thermal contraction during cooling is released by the abrupt onset of step bunching in the foil at ~1030 K, triggering the formation of graphene folds perpendicular to the step edge direction.) By growing below this temperature, we achieved fold-free, adlayer free, large area single crystal graphene films. The resulting films showed highly uniform transport properties: field-effect transistors prepared from these films exhibited average room temperature carrier mobilities of around $7.0 \pm 1.0 \times 10^3$ $cm^2V^{-1}s^{-1}$ for both holes and electrons. The process is also scalable, permitting simultaneous growth of graphene of the same quality on multiple foils stacked in parallel. After electrochemical transfer of the graphene films from the foils, the foils themselves can be reused essentially indefinitely for further graphene growth. This work is published (*Nature*, 2021, 596(7873): 519-524; M. Wang, M. Huang, D. Luo, Y. Li, M. Choe, W. K. Seong, M. Kim, S. Jin, M. Wang, S. Chatterjee, Y. Kwon, Z. Lee, R. S. Ruoff). *We appreciate support from the Institute for Basic Science (IBS-R019-D1).*

Figures

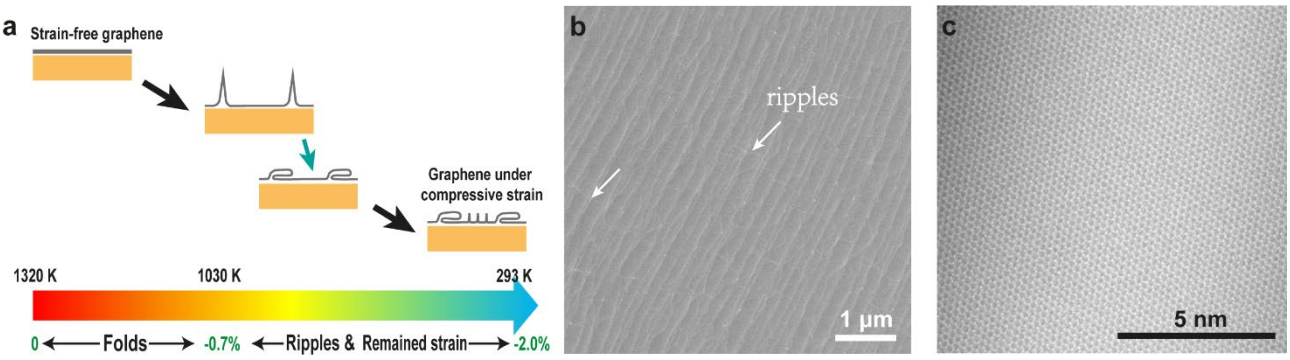

Figure 1: (a) Schematic of formation mechanism of graphene folds. (b, c) SEM image (b) and HR-TEM image (c) of fold-free single crystal single layer graphene films grown on Cu-Ni(111) foils with 20 at.% Ni.