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MS-2-O-2415 Evidence of re-crystallization in graphene probed by atomic scale imaging

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Graphene shows great potential for future nanoelectronics due to its extraordinary electronic properties and structure-engineerable nature. Recently CVD based graphene production technology has given insights to the possibility of large scale application. Since the CVD grown films are typically polycrystalline with numerous grain boundaries (GBs), it is important to characterize and control grain size and GBs, generally believed to limit the transport properties of graphene film. In this work, we report on a peculiar grain evolution occurring during the growth process. This evolution is revealed by means of Raman spectroscopy that gives access to a statistically characterization of the graphene structure (grain size, presence of defects etc.). Besides, we need some other complementary techniques to fully understand the detailed atomic structures. This is done by means of atomic-resolution TEM. Indeed, following the invention of aberration corrector (AC), HRTEM direct imaging has become possible on one atom thick layer of carbon. This technique allows us to analyse the detailed atomic structures of and around the GBs together with fundamental information of each grain. In this work, graphene continuous films synthesized on platinum substrate with a specific configuration of CVD set-up are atomically characterized using AC-HRTEM imaging. Fig. 1 shows Raman spectra with corresponding HR-TEM images of our samples at different stages of the growth process. The evolution of both orientation and size of the grains is observed during the process. Nanometer size grains already connected with various orientations (see FFT on the inset) at stage I are further re-oriented and merged together along some pre-dominant directions in stage II and finally form large single crystal domains in the last stage. In stage II, two neighboring grains are typically aligned zig-zag to armchair at the boundary (Fig. 2a), which might be a low-energy crystallographic configuration. Slightly misoriented neighboring domains are connected with the presence of some dislocations (Fig. 2b). A lot of small domains are observed enclosed within larger ones (Fig. 2c) with the same zig-zag to armchair misorientation. Generally in CVD processes, the orientation and achievable size of grains are determined at the early nucleation stage. In our case, the small grains already form a continuous film at the early stage and further transform to low defective large crystals. We infer that such large scale re-crystallization of graphene is enhanced thanks to the platinum substrate. The latter hypothesis is supported density functional theory (DFT) calculation.

[1] O. Lehtinen et al., Nature Communications, DOI : 10.1038 /ncomms3098 (2013)

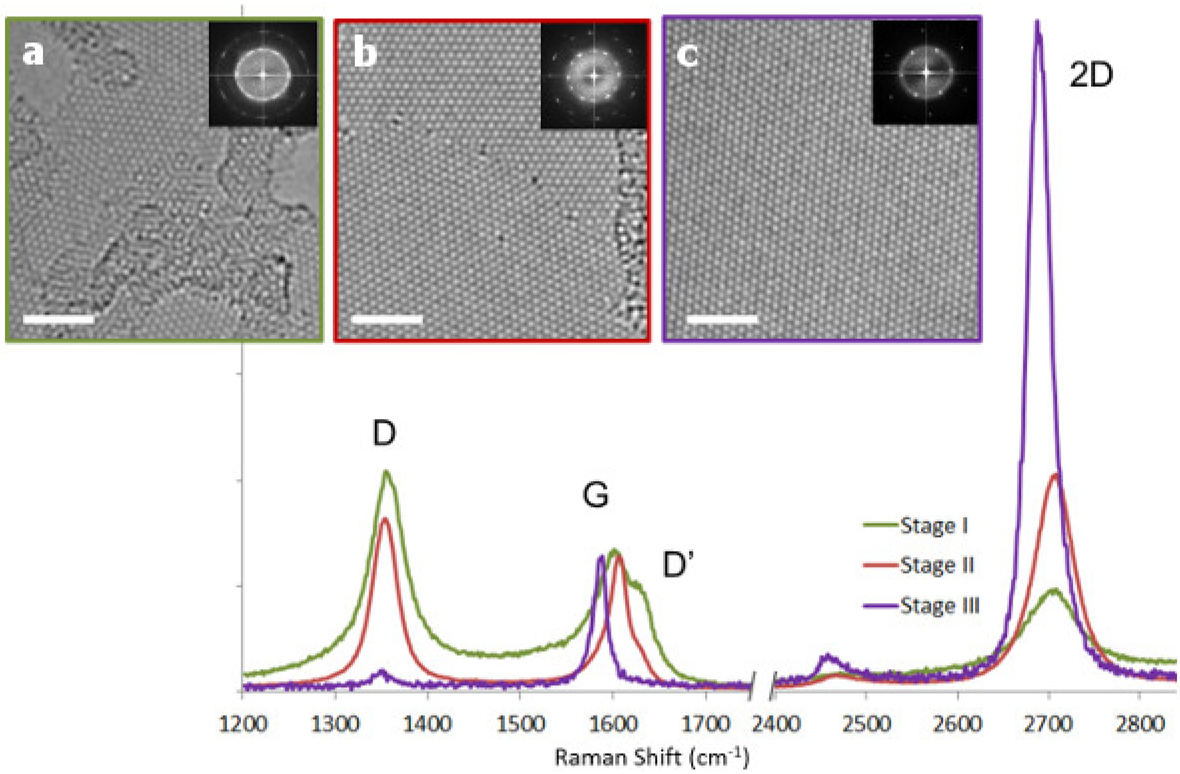

Fig. 1: Raman spectra of graphene continuous films at different stages of the synthesis process. HR-TEM images (a-c) correspond to typical atomic structures of stage I, II and III, respectively. Fourier transform shown on insets is taken from 50x50nm^2 area of each sample. Scale bar is 2 nm.

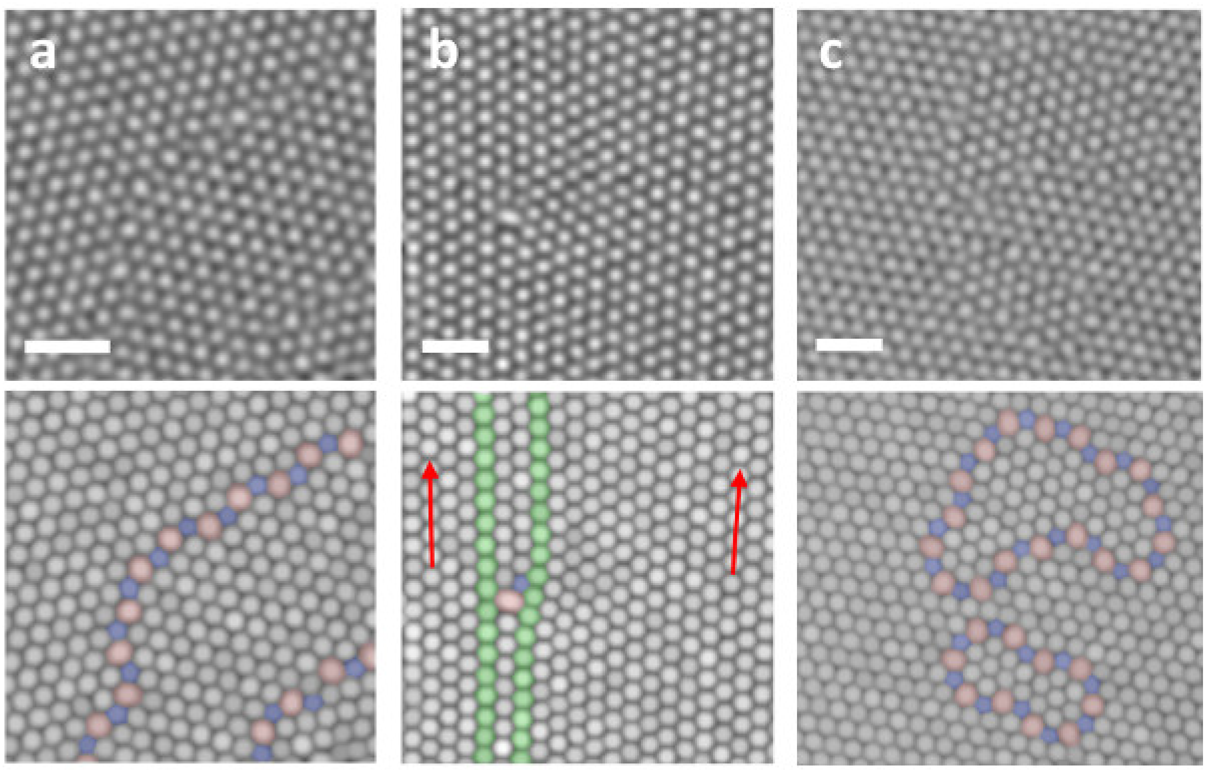

Fig. 2: The upper panels show low-pass Fourier filtered images and the lower panels with maximum filtering [1]. Typical structures observed at stage II of (a) boundary between grains aligned zigzag to armchair, (b) dislocation between two grains with slight angle mismatch and (c) small domains inside another large grain. Scale bar is 1 nm.