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MS-2-P-1822 Relations between deficiencies in CVD deposited graphene and the lattice defects of the Ni (111) substrate

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In this work graphene layers were investigated that were deposited by chemical vapor deposition (CVD) on nickel (111) thin film substrates. The Ni (111) thin substrates themselves were grown previously on bulk sapphire (0001) substrates at 550°C with 33nm/min. growth velocity. The nickel layer grew epitaxially on the sapphire, as it is shown by the diffraction in figure 1. The Ni (111) planes were parallel to the sapphire (0001) planes. Twin boundaries were formed in the nickel layers. At some places grains were rotated with 30° about an axis perpendicular to the sapphire surface.
The graphene deposited on the Ni (111) was investigated by TEM, Cs-corrected HRTEM, STM, AFM and by SEM / EBSD. According to [1] above 600°C, attempts to grow graphene on Ni (111) only resulted in the appearance of multi-layer turbostratic graphite. Here we show how the orientation of the first graphene layer and the appearance of additional turbostratic layers are related to the quality and orientation of the Ni (111) substrate. The nickel substrates were annealed in hydrogen before the graphene deposition. The graphene was deposited at atmospheric pressure from a mixture of argon, methane and hydrogen gases. The sample was cooled in argon and hydrogen gas mixture. Large area continuous single-layer graphene formed on top of nickel (111). Above it both another turbostratic graphene partial layer (Fig. 2), and graphite flakes were formed at certain locations. Such flakes are seen in the SEM image of figure 3.a. The continuous single-layer graphene formed epitaxially on the nickel (111) and the epitaxial relation was corroborated by both STM and TEM analyzes.
The growth of the thinner and smaller flakes was suppressed by changing the gas concentration. SEM and EBSD studies showed that the thicker flakes grew above the incoherent twin boundaries and high-energy (30°) grain boundaries of nickel.
Our results show that the first atomic layer grew as a continuous and epitaxial graphene layer on nickel (111). The additional local turbostratic layers and graphite flakes grew above the incoherent twin boundaries or high-energy boundaries in the nickel substrate. No thicker flakes were observed above coherent twin boundaries.

[1] Patera L.L. et al., ACS Nano 7 (9) 7901–7912 (2013)

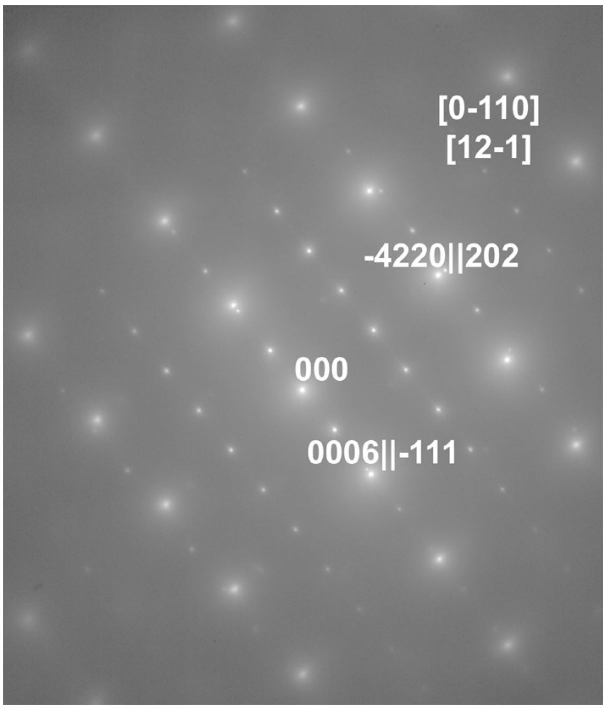

Fig. 1: SAED diffraction pattern recorded from the nickel and sapphire interface. The nickel layer grew epitaxially on the sapphire.

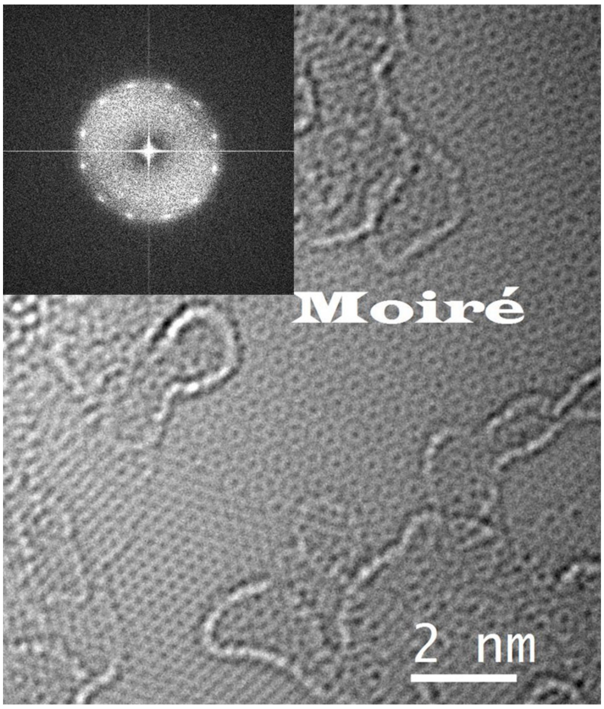

Fig. 2: Cs-corrected HRTEM image recorded from a local turbostratic graphene flake over the large area continuous graphene layer.

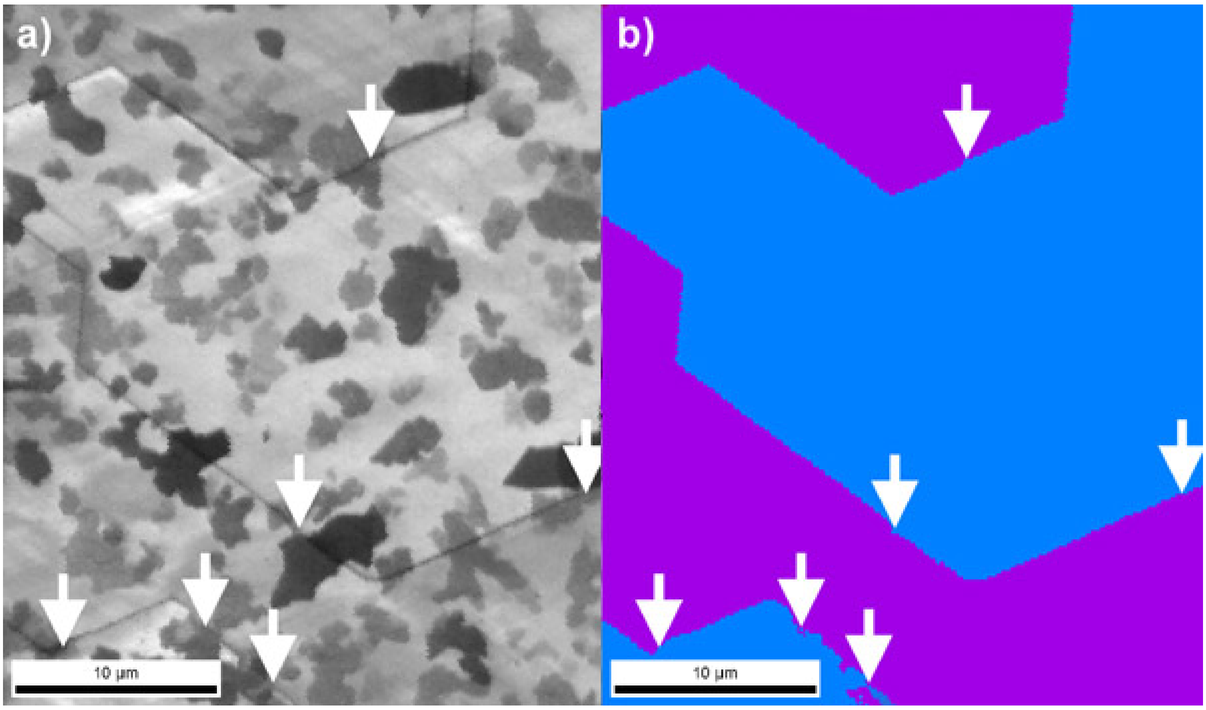

Fig. 3: Figure a) SEM image. The dark patches of different gray shades are from an additional turbostratic graphene layer and graphite flakes. Figure b) EBSD orientation map. The straight lines mark locations where twin boundaries reach the surface of nickel.