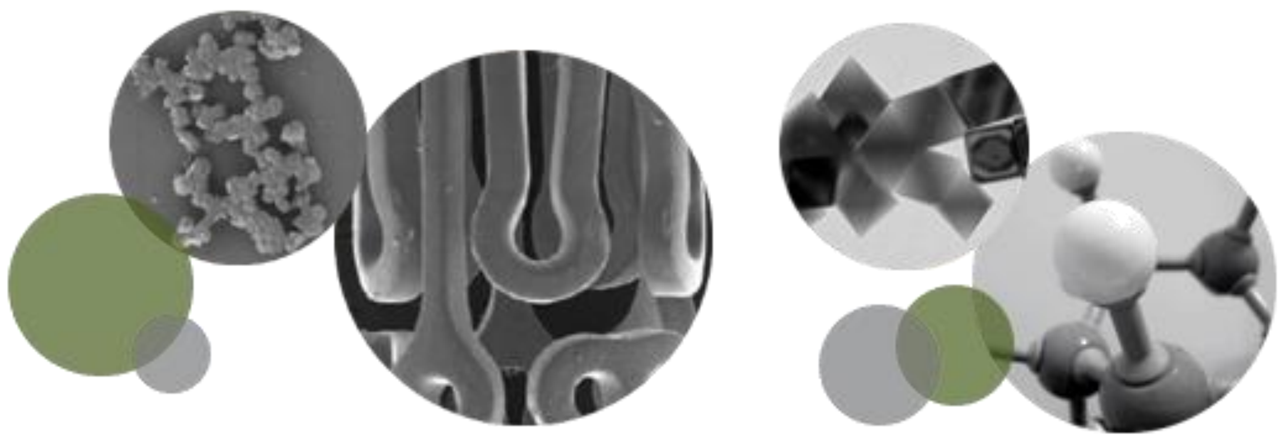
2020 nanoPortugal CONFERENCE ONLINE

September 23-24, 2020



Nanoscience and Nanotechnology International Conference

Oxygen assisted monocrystalline graphene growth by chemical vapor deposition

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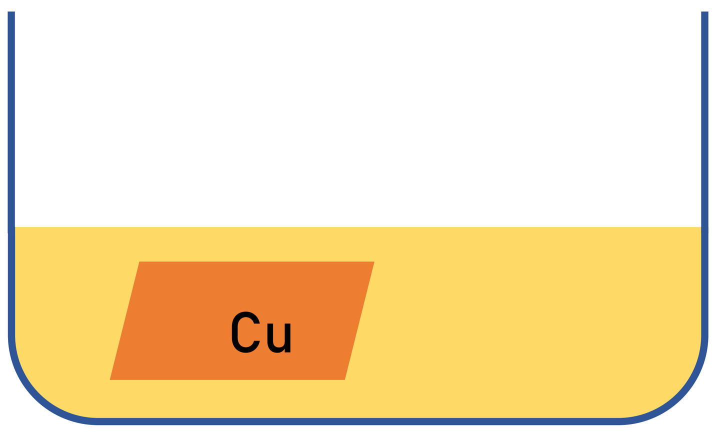
Motivation

Monocrystalline single layer graphene is an appealing material for applied research due to its exceptionally large carrier mobility - a key parameter for most applications in electronics - and strong THz optical response, for photonic applications. However, for an industrial scale exploitation of these properties, it is necessary to obtain high quality graphene by scalable means, e.g., by chemical vapour deposition (CVD) [1]. For single layer graphene growth it is preferable to use copper instead of, for example, nickel substrates due to the low carbon solubility in copper [2]. The number and size of the graphene crystal grains is, in a first approximation, determined by the copper substrate characteristics. Therefore, it is necessary to treat the high-purity copper for control of its cleanliness, oxidation state, and roughness. . Substrates were treated in an ultrasonic bath with an acidic solution of HCl and $FeCl_3$, followed by oxidation in air (hot plate) at 180°C for 30 minutes. Keeping an extended oxidized surface is key to obtain very low nucleation density, providing graphene crystals of large size and good quality. For graphene growth, the copper substrates were enclosed in a graphite confinement box to increase growth rate [3] while protecting the sample from silicon oxide and other contaminations coming from reactions with the quartz walls of the reactor. A secondary height-controlled sapphire cavity is used to accommodate the substrate inside the primary graphite cavity and release in situ trace amounts of oxygen that keep the Cu substrate oxidation level, and further increase growth rate and reduce nucleation density. The consistency in this step is fundamental to achieve a good reproducibility of the results. Samples are characterized by microscope inspection for flake size and morphology, after which they are transferred onto Si/SiO_2 substrates, using the wet polymer transfer process, for Raman spectroscopy analysis.

Graphene Flakes Deposition Steps

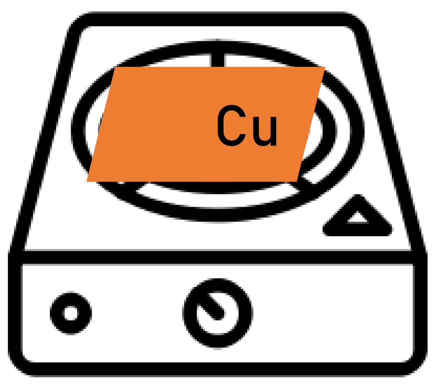
Substrate preparation

Treatment with acidic solution to reduce copper foil roughness at a microscopic level.



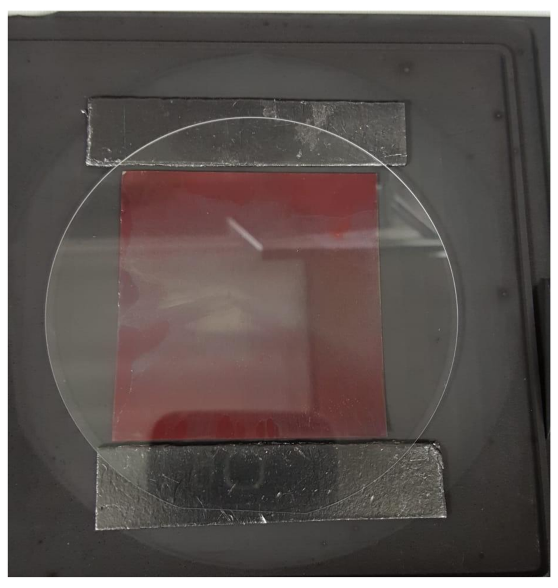
Ultrasound bath in solution of $FeCl_3$, HCl, H_2O (~1 min)

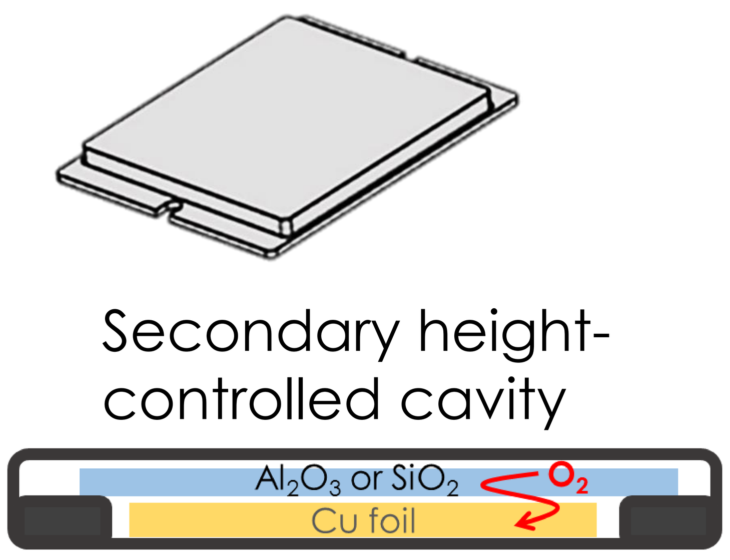
Pre-deposition oxidation of the substrate using a hot plate at 180 °C for 30 min.



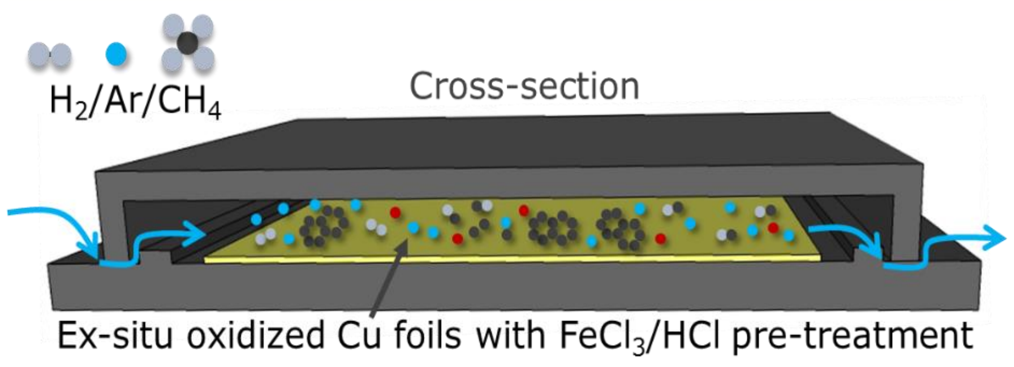
CVD Growth

Mounting of the substrate and sapphire disk in a graphite box using graphite spacers. The ensemble is loaded into a reactor.



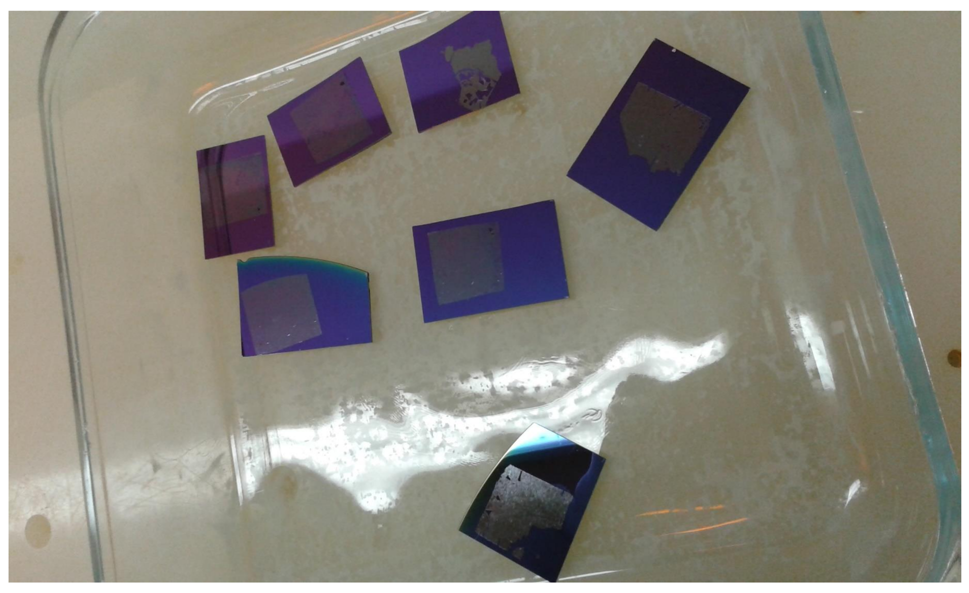


Annealing for 30 min in an Argon atmosphere.
Injection of CH_4, H_2, and Ar at high temperature and low pressure.

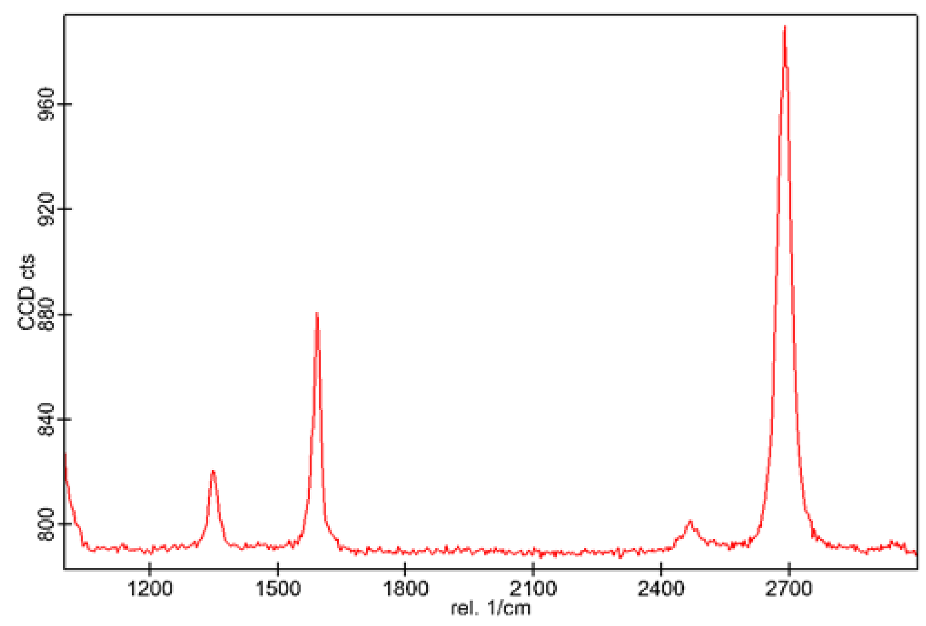


Characterization

Transferring of graphene flakes samples onto Si/SiO_2 substrates via the polymer assisted wet transfer method.

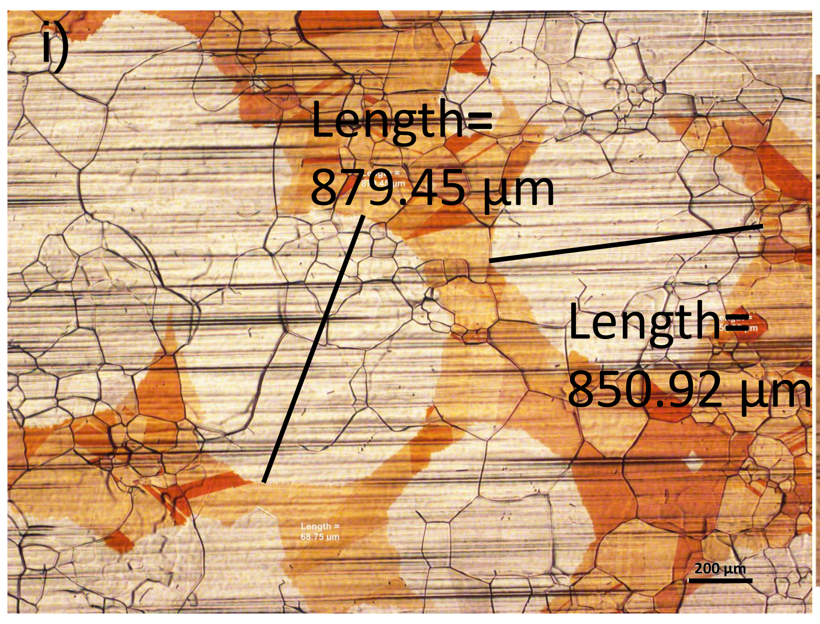


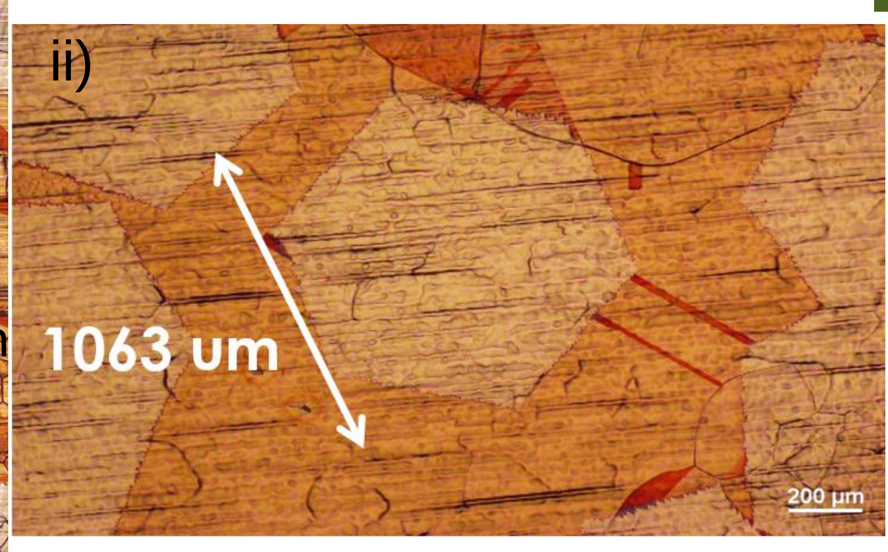
Raman spectroscopy to analyse the quality of the material.



Summary

A process for graphene flakes growth was presented. The important parameters such as the substrate preparation, CVD growth, and the transfer were addressed. The quality of the graphene flakes was evaluated through the Raman spectrum after the transfer.





Legend

Graphene Flakes on copper foil grown under the same CVD conditions. **i)** Round flakes with over 850 µm diameter. **ii)** Large hexagonal flakes with over 1.5 mm diameter.

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