Growing Twisted Bilayer Graphene using Chemical Vapour Deposition

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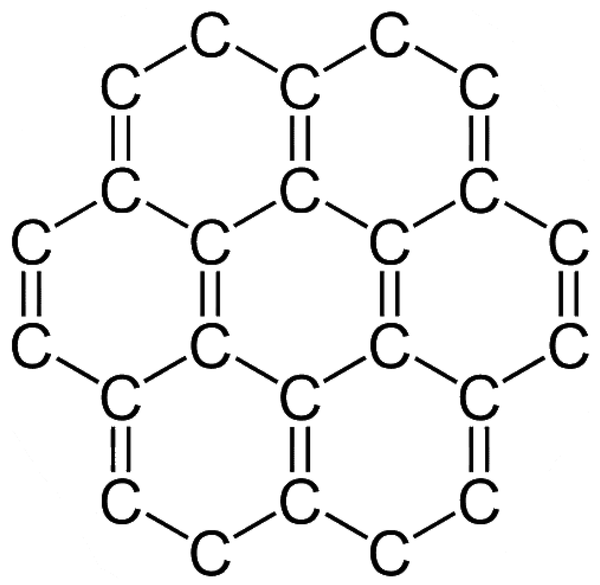
What is Graphene?

Graphene is a one atom thick molecule made up of carbon atoms arranged in a hexagonal lattice. Why is graphene of interest? Well, its not exactly graphene, but rather twisted bilayer graphene which is of interest to us for its many unique properties.



The chemical structure of graphene

Twisted Bilayer Graphene

When growing graphene, a second layer can begin to grow beneath it, thus forming a graphene bilayer. As the second layer forms however, it can grow in a rotated orientation compared to the top layer of graphene, thus causing a twisting angle between the two layers.

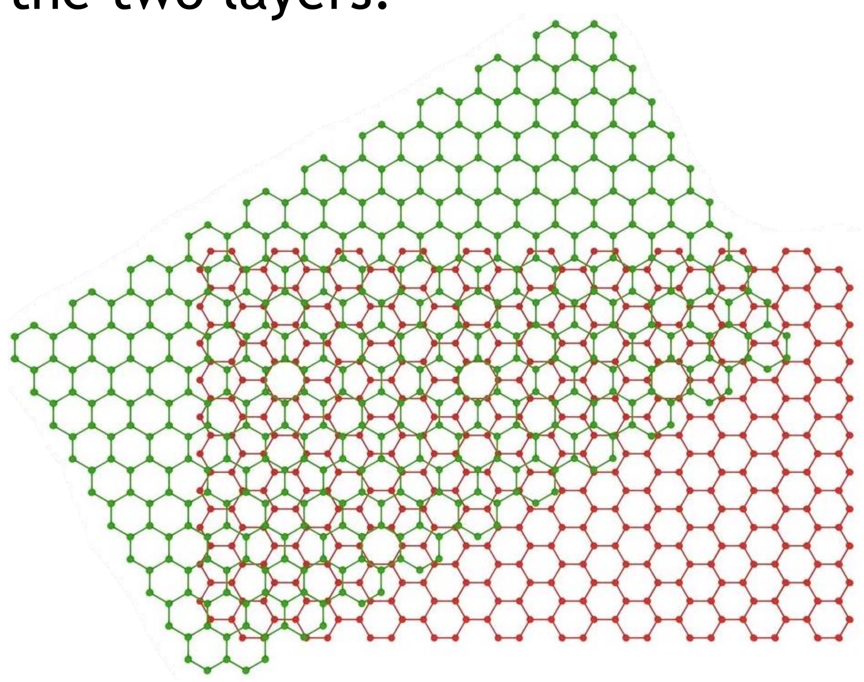


Diagram demonstrating the twisting angle between 2 layers of graphene

At different twisting angles, the bilayer graphene can exhibit different properties. One such angle of interest lies around 1°, and causes the bilayer to have non-linear optical properties, which is of interest for quantum computing. Other twist angles have also been shown to cause superconductive effects in the graphene.

Acknowledgements

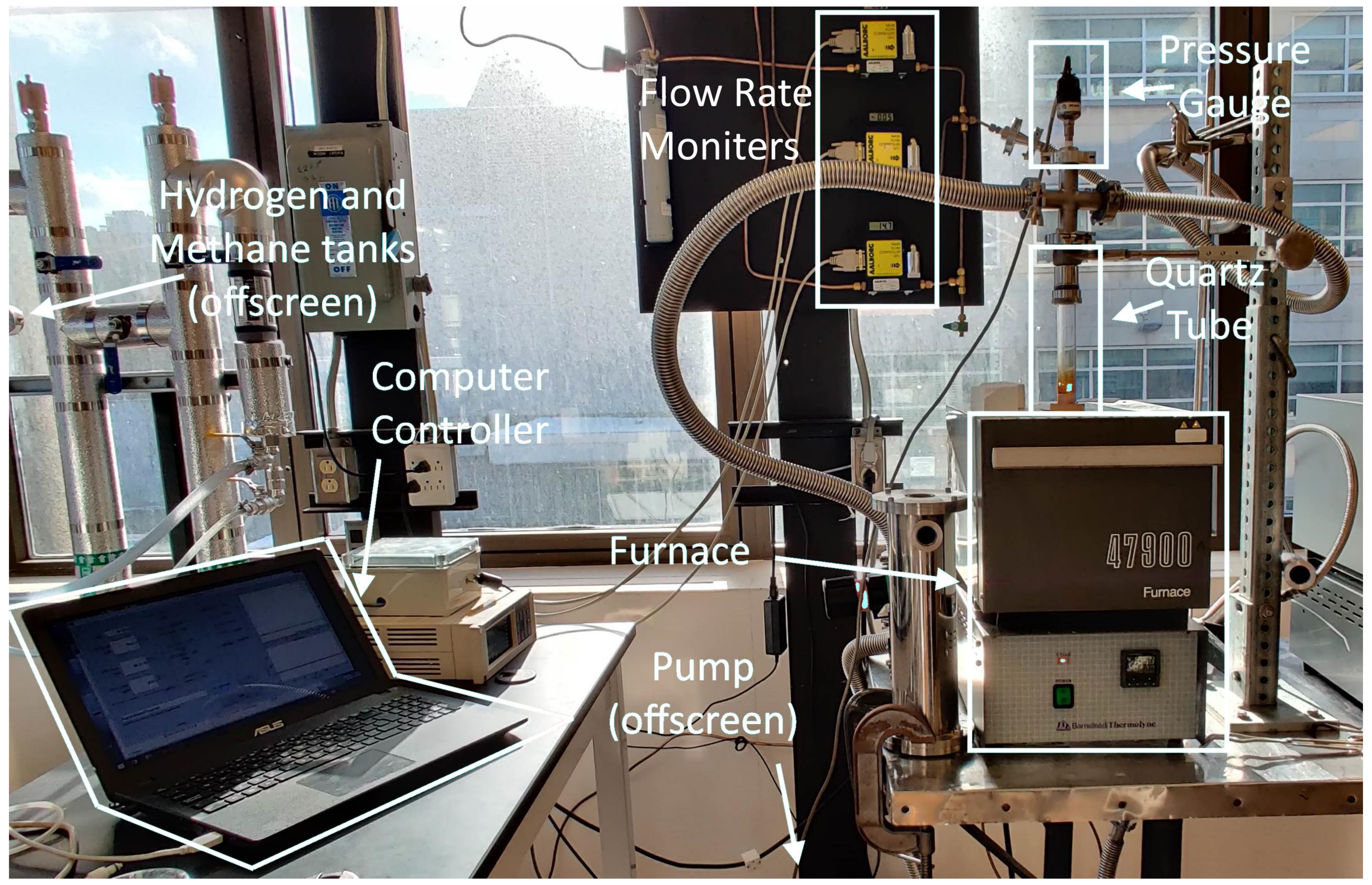
Supervisor: Michael Hilke (*hilke@physics.mcgill.ca*)
Lab Partner: Nicolas Delnour

Growing Bilayer Graphene

The method used to grow bilayer graphene is known as chemical vapour deposition (CVD). In CVD, a gas is passed through a heated system where elements of the gas get deposited onto a substrate. In our case, we use copper as the substrate and hydrogen and methane as our gases. The equation for this deposition is:

$$3CuO + CH_4 + H_2 \rightarrow C + 3Cu + 3H_2O.$$

The carbon in methane is deposited on the copper in order to form graphene on its surface. While hydrogen doesn't play a direct role in the formation of graphene, it acts as an etchant to reduce the nucleation density of new crystals and to keep the copper surface clean. The CVD system can be seen below.

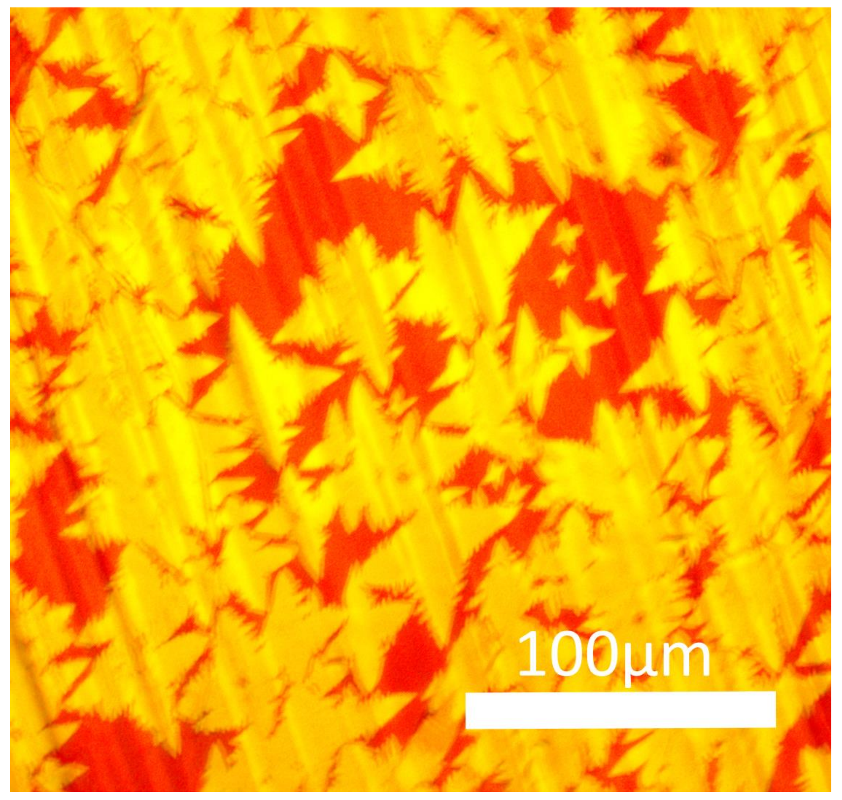


An annotated image of the chemical vapour deposition system. Shown in the image is the furnace, the quartz tube, the pressure gauge, the flow rate monitors and the computer controller. The pump and gas tanks were not able to be captured in the image.

The size of a graphene bilayer is correlated with the size of the graphene crystal it is formed under, and so to optimize its size we want to optimize the size of each graphene crystal. This is done by varying the many different parameters that go into the growth of graphene, notable the annealing time for the copper substrate, the purity of the copper, the temperature of the furnace, the concentration and pressure of hydrogen and methane and the amount of time the graphene layers are allowed to grow, to name a few.
The goal is to determine the perfect parameters in order to grow the largest graphene crystals, and therefore bilayers.

Observing the Crystals

After undergoing CVD, the copper sample is placed on a hot plate. This is because the heat causes the bare copper to oxidize and change colour, whereas the areas covered by graphene are protected from oxidation. By doing this, these single atom thick graphene molecules become visible!
The graphene forms a layer of crystals which is categorized by the size of the crystals, their shape, their dendricity (or spiky-ness) and the nucleation density of the crystals.

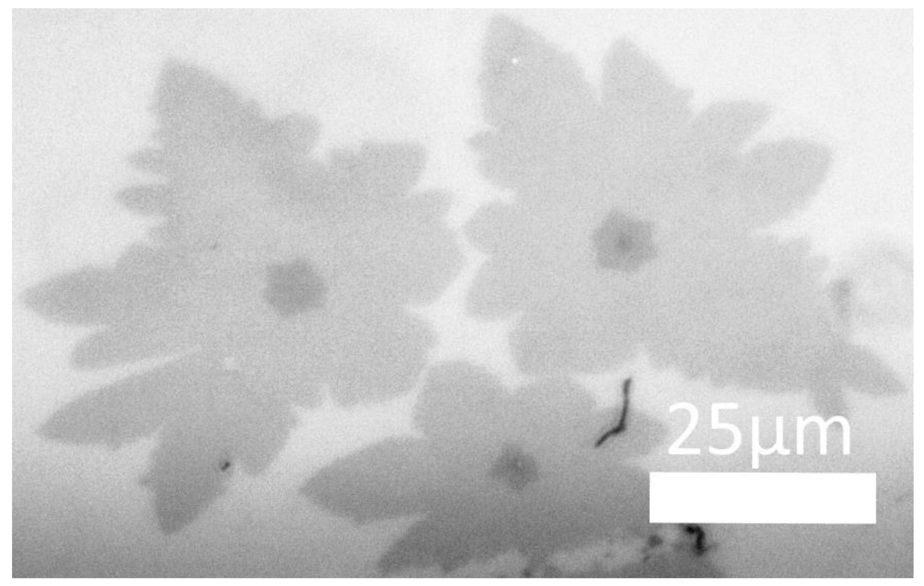


Graphene crystals grown on copper foil

The higher the nucleation density, the more crystals, but the smaller they are. Therefore, in order to observe a lot of large graphene bilayers, a balance has to be found.

Observing the bilayer

To be able to finally view the bilayers underneath the graphene crystals, the graphene first needs to be transferred from the copper substrate to a silicone substrate. Once this is done, the bilayers can be viewed under a microscope as the dark area at the center of each graphene crystal. On some crystals, such as the one in the top right of the above image, an even darker zone can be seen at the nucleation site of the crystal, indicating the presence of a third layer of graphene formed underneath the second layer.



Three graphene crystals with visible bilayers. The colour, brightness and contrast of the image were altered in order to better view the graphene bilayers

What about the twist angle?

In order to measure the twist angle between the overlaying and underlying layers of graphene, the sample needs to be observed using Raman spectroscopy or a scanning electron microscope (SEM), which has not yet been done. Therefore, to observe the unique properties of twisted bilayer graphene, there is still work to be done!