Graphene Oxide (GO) and reduced Graphene Oxide (rGO): Electronic properties

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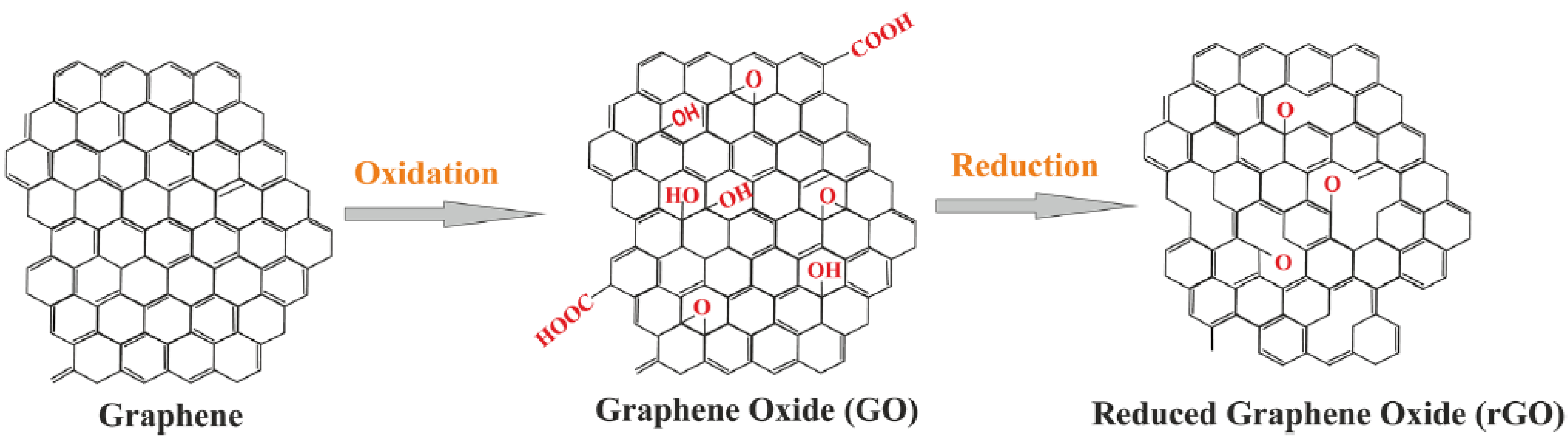
Structure of GO and rGO

GO

- Two-dimensional, disordered and heterogeneous material
- Sp^2 and sp^3 hybridisation

rGO

- Some of the functional groups removed.



Adapted from https://doi.org/10.1007/s40097-018-0265-6

Electronic properties of GO and rGO

GO

Electron conduction occurs through hopping.

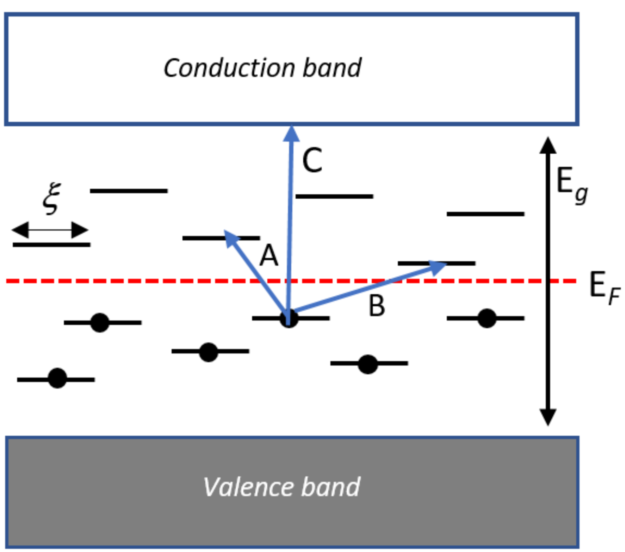
The energy gap depends directly on the fraction of sp^2/sp^3 hybridized domains.

rGO

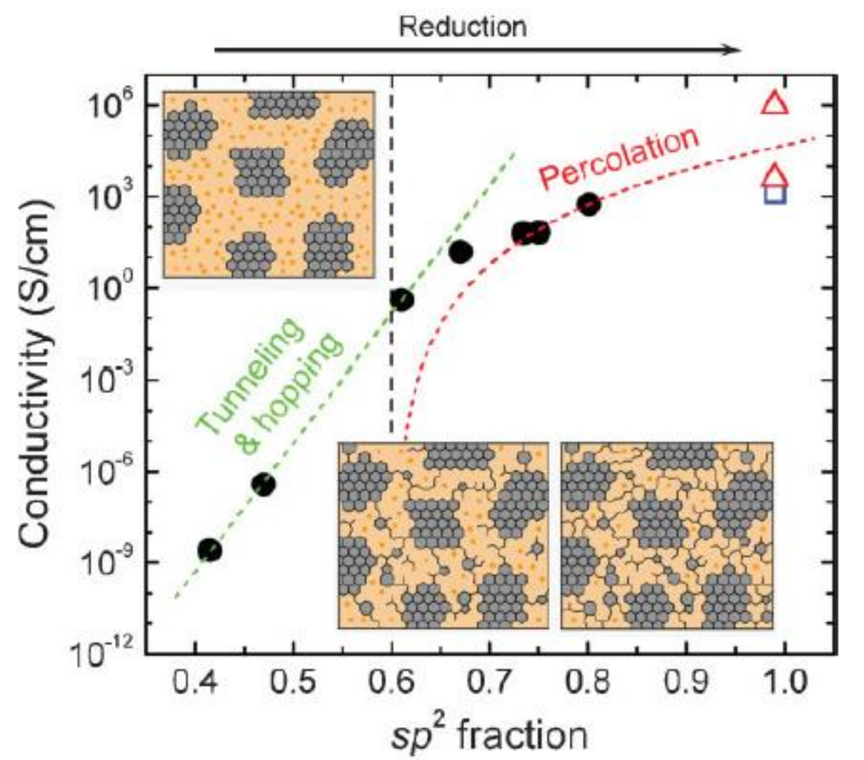
New sp^2 clusters are created.

It enhances conductivity through hopping or even through percolation if it is highly reduced.

$$\Gamma_{i,j} \propto \exp\left(-\frac{\Delta E_{i,j}}{kt}\right)\exp\left(-\frac{2r_{i,j}}{\xi}\right)$$

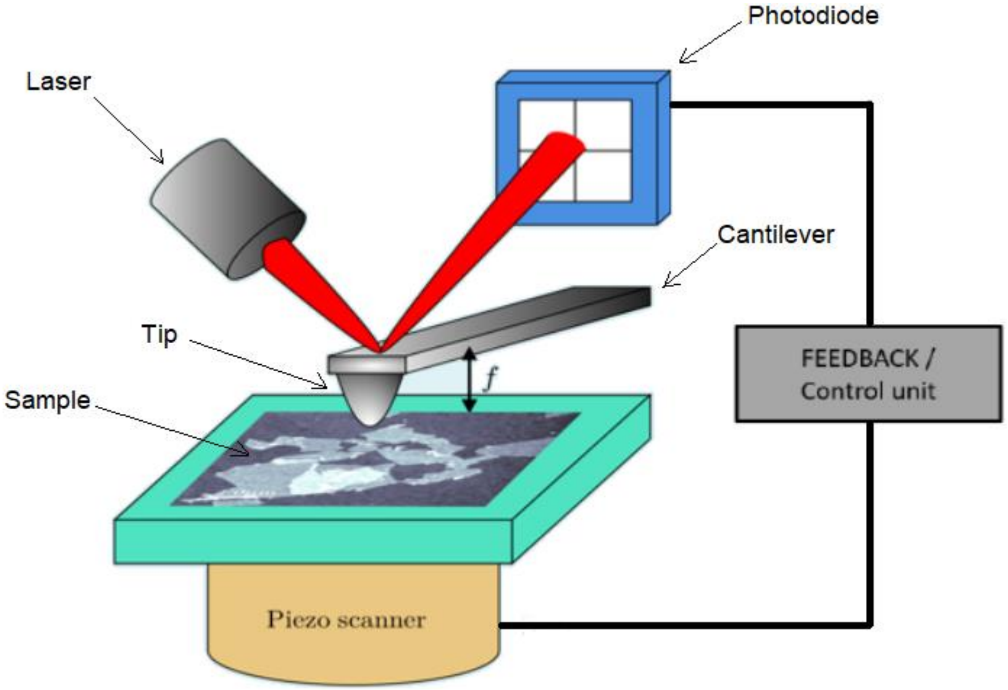


Adapted from M. Pollak, M. Ortuño, and A. Frydman, The electron glass. Cambridge University Press, 2013.



Adapted from https://*doi.org/10.1002/adma.200903689*

Atomic Force Microscope and KPFM



External voltage applied

$$V_{bias} = V_{\mathrm{DC}} + V_{\mathrm{AC}} \sin(\omega_{\mathrm{elec}} t)$$

$$\Delta f = \underbrace{-\frac{f_0}{2k_{cantilever}} \frac{\partial^2 C(z,\theta_i)}{\partial z^2} \left[\frac{V_{ac}^2}{4} + \frac{1}{2}(V_{DC} - V_{SP})^2 \right]}_{\Delta f_{DC}}$$

DC signal

$$\underbrace{-\frac{f_0}{2k_{cantilever}} \frac{\partial^2 C(z,\theta_i)}{\partial z^2} (V_{DC} - V_{SP}) V_{ac}}_{\Delta f_{\omega}} sin(\omega t)$$

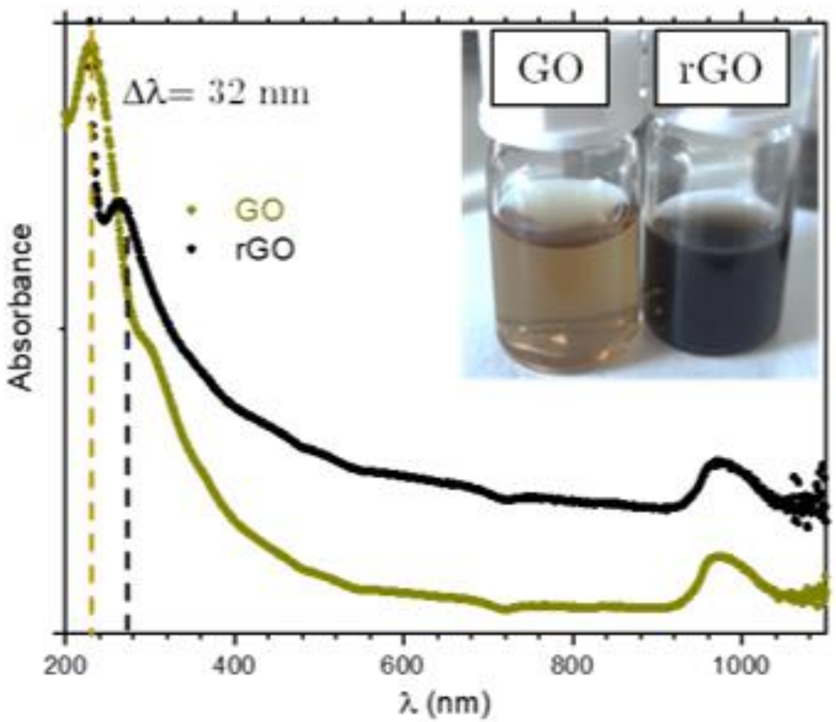
$V_{DC} = -V_{SP}$ allows to map the surface potential (KPFM)

$$+ \underbrace{\frac{f_0}{2k_{cantilever}} \frac{\partial^2 C(z,\theta_i)}{\partial z^2} \frac{V_{ac}^2}{4}}_{\Delta f_{2\omega}} cos(2\omega t)$$

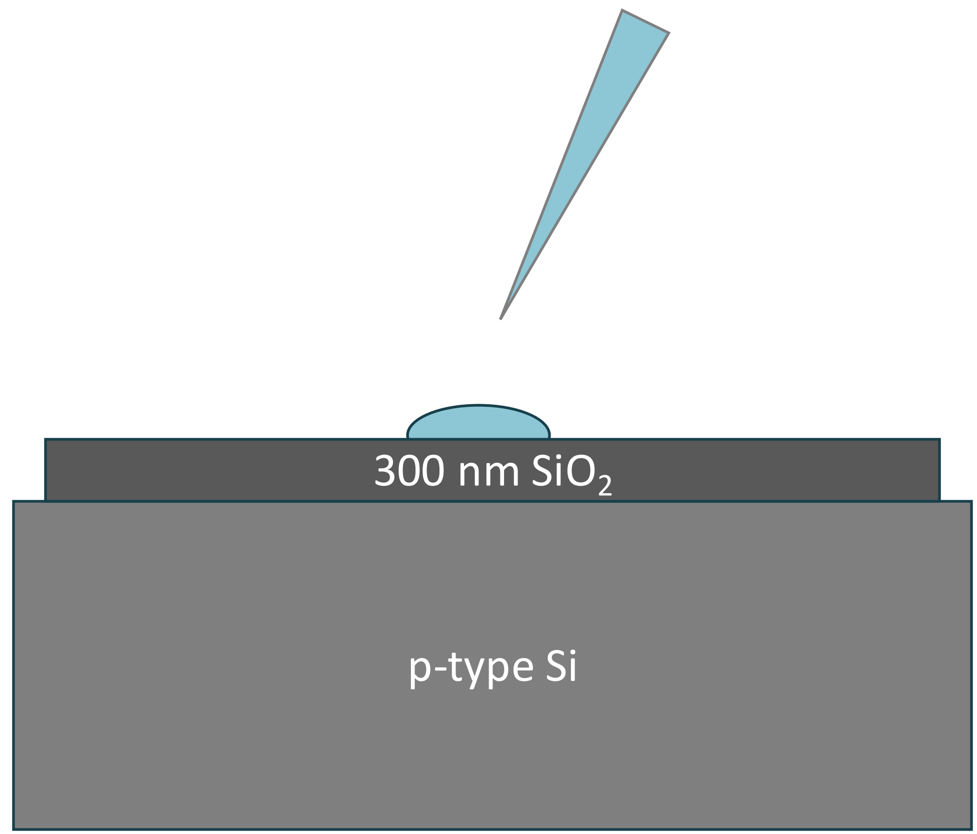
Information about the tip-sample capacitance

Preparation of the sample

Dispersion of GO and rGO in water.

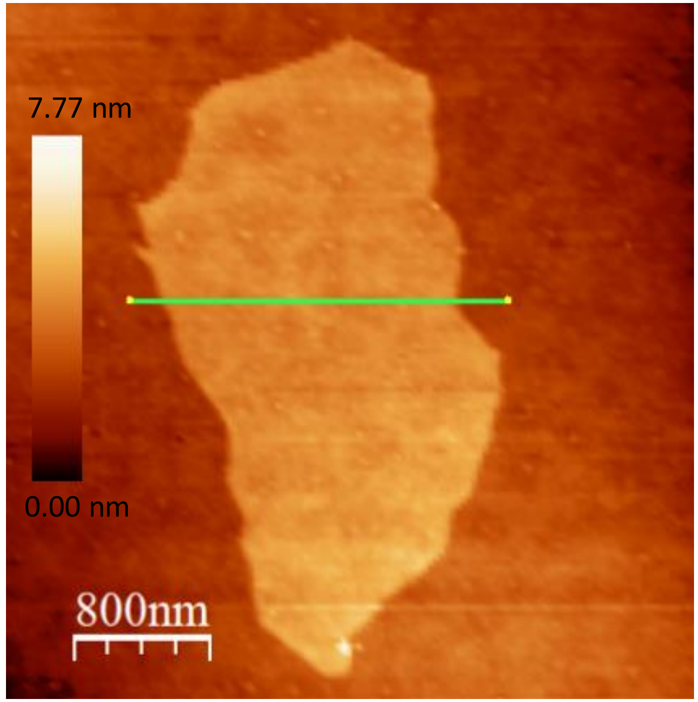


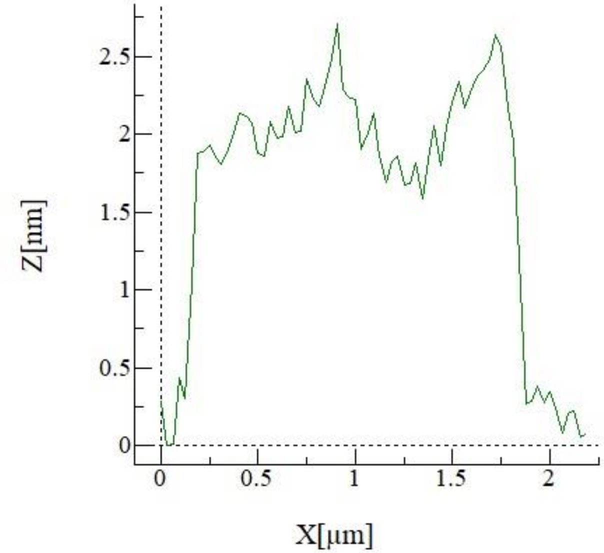
Drop-casting



GO flake

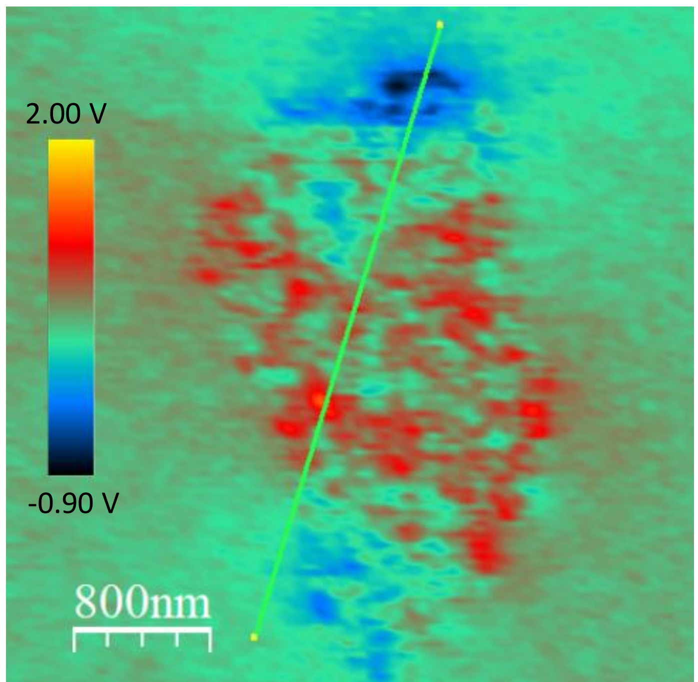
Topography:
Average height of the GO flake is approximately 2 nm.

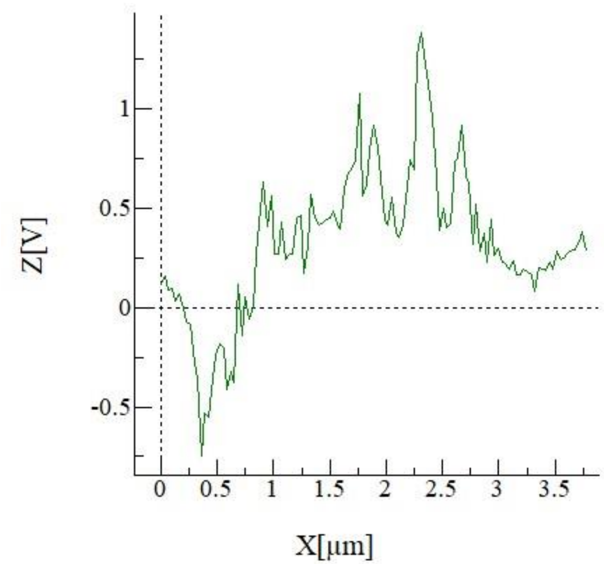




KPFM:
Surface potential varies among different regions.

Localized charge domains.

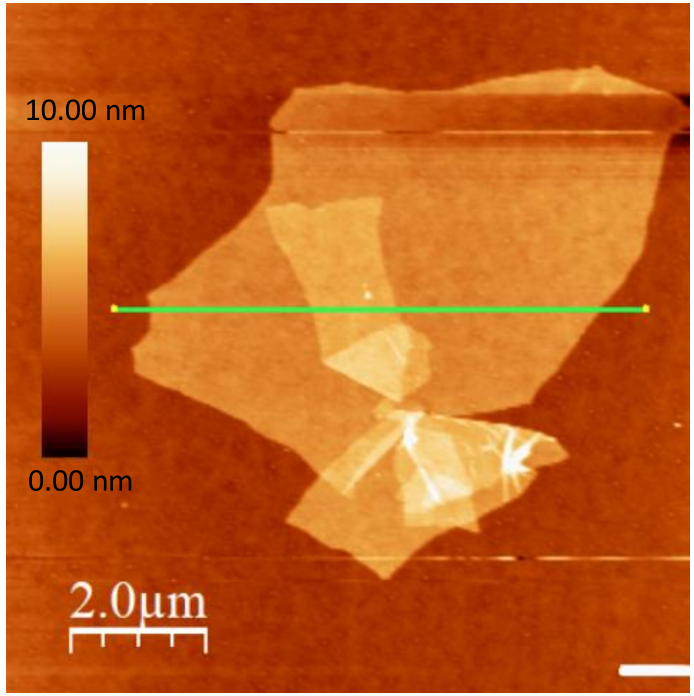


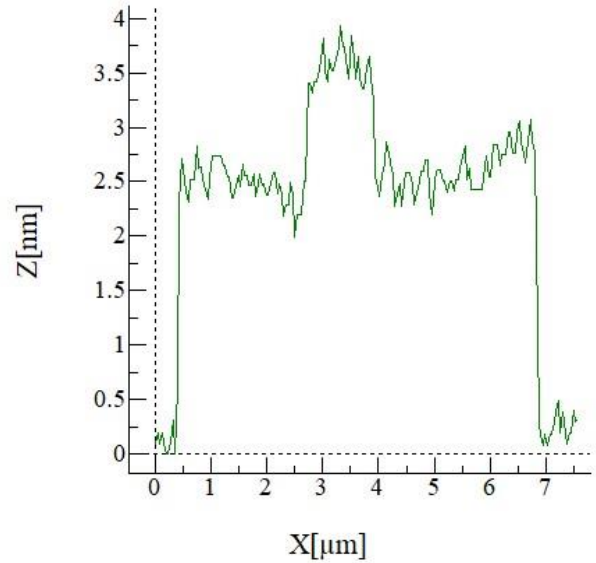


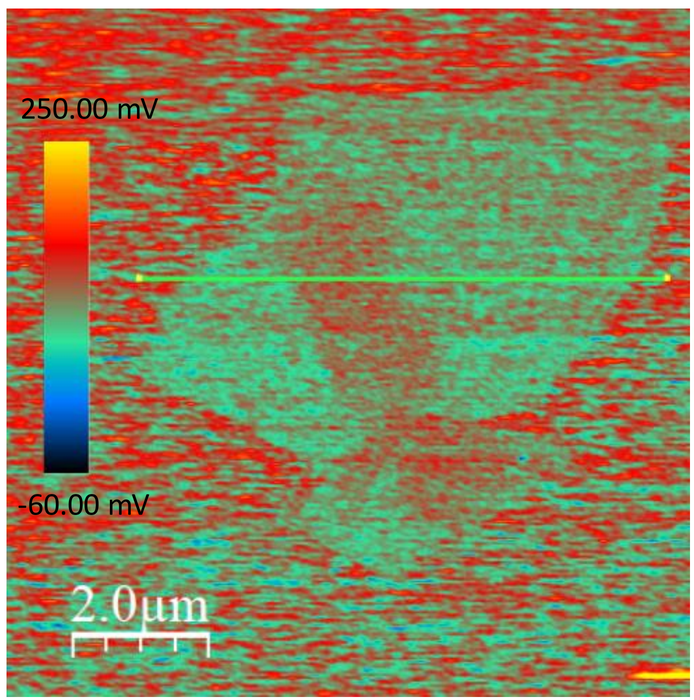
rGO flakes

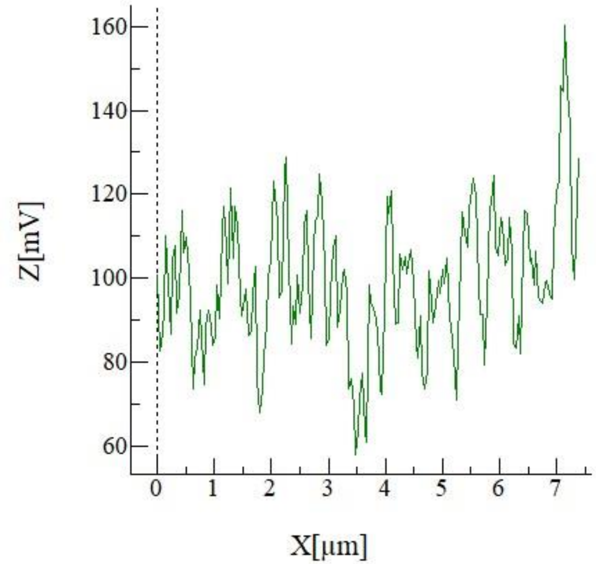
Topography:
Average height of the rGO flakes is approximately 2.5 nm. The double layer has a total height of 3.75 nm.

KPFM:
The surface potential in rGO is more homogeneous compared to GO.









Charge injection in GO and rGO

Electrical discharge of approximately 30 ms.

GO: The charge is highly concentrated in a small region.

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Insulator

rGO: The charge has spread faster than the acquisition time.

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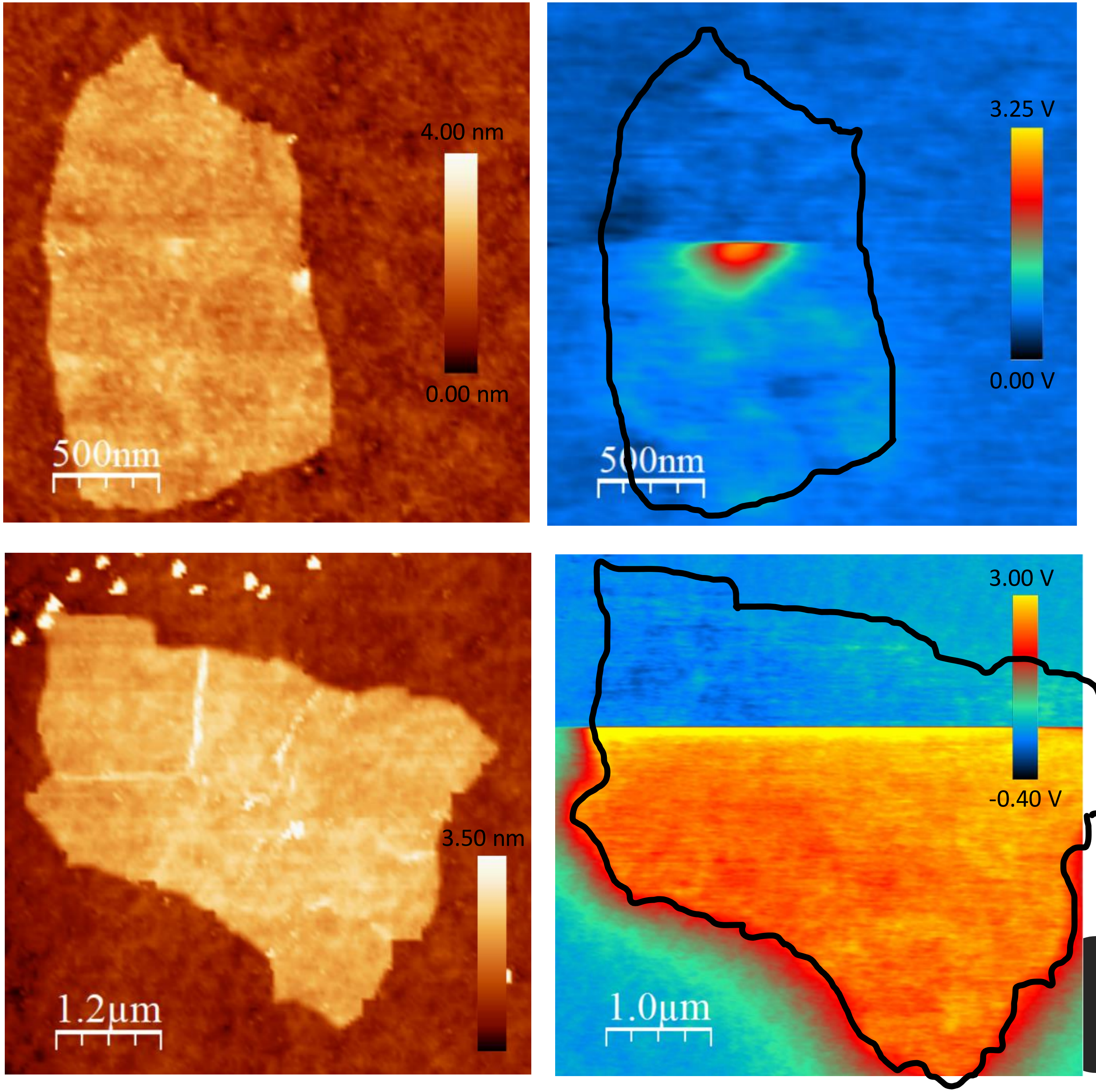
Higher conductivity

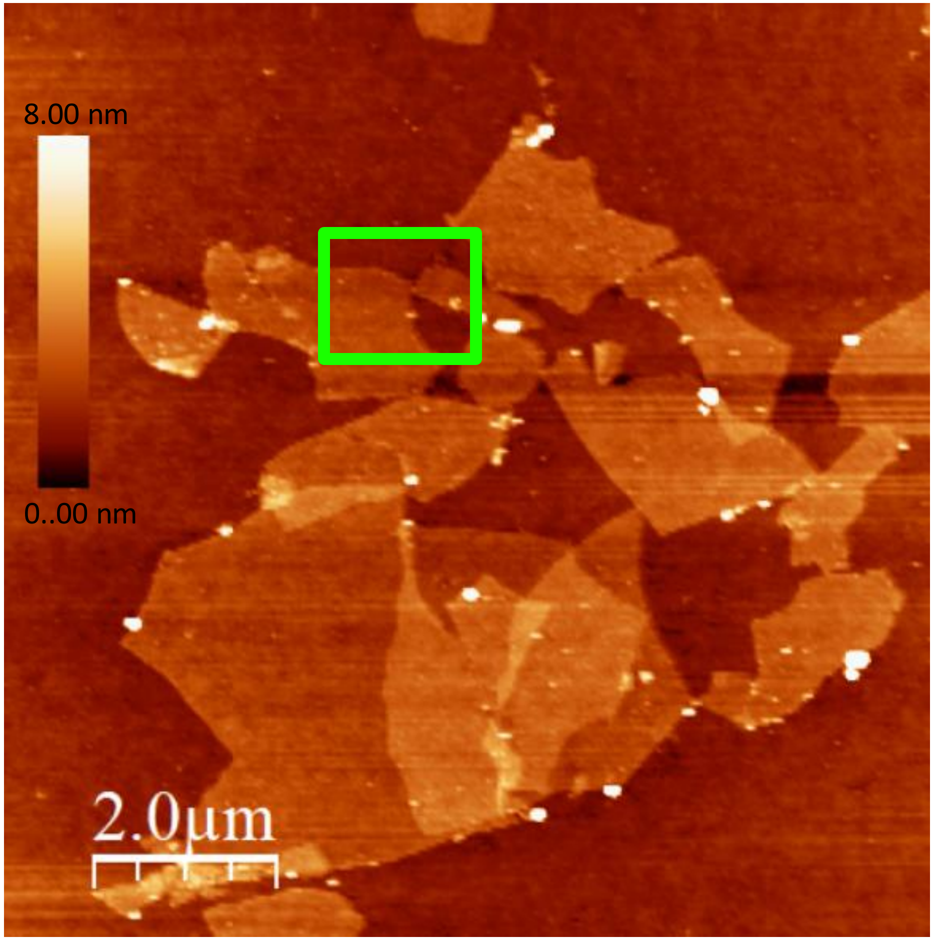


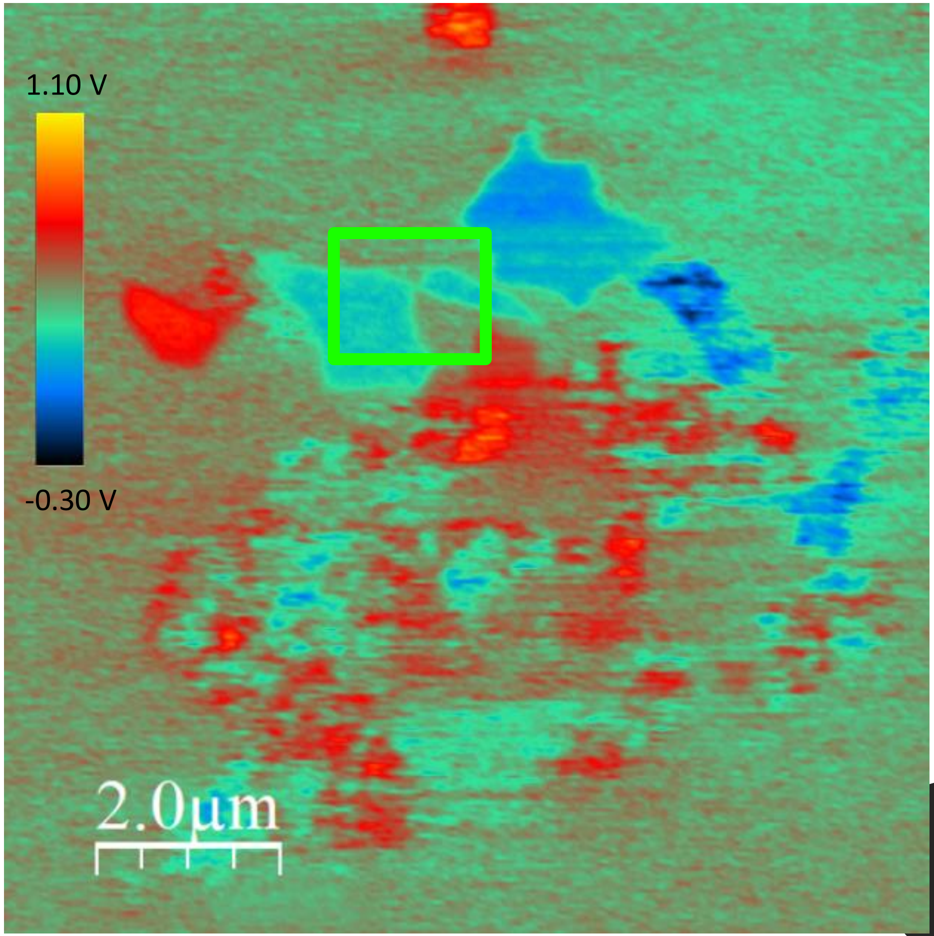
Comparison between co-deposited GO and rGO

Topography → GO and rGO cannot be distinguished.

KPFM → rGO: More uniform surface potential

GO: Heterogeneous charge distribution.





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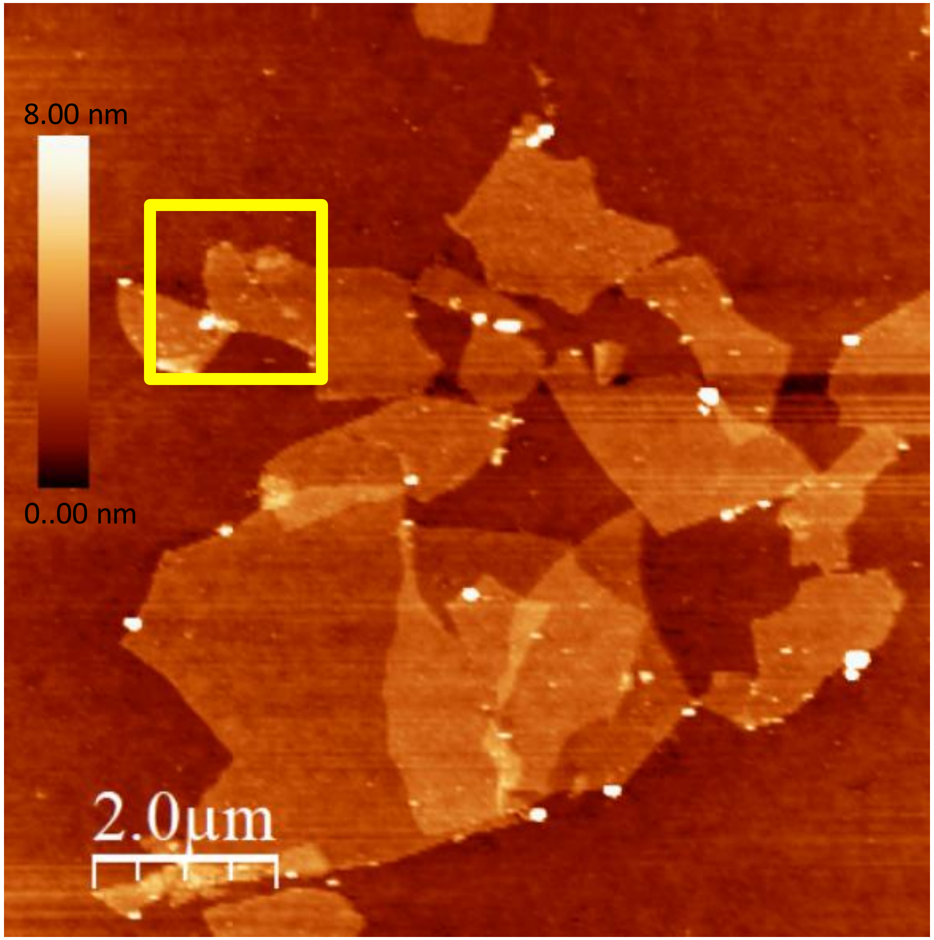

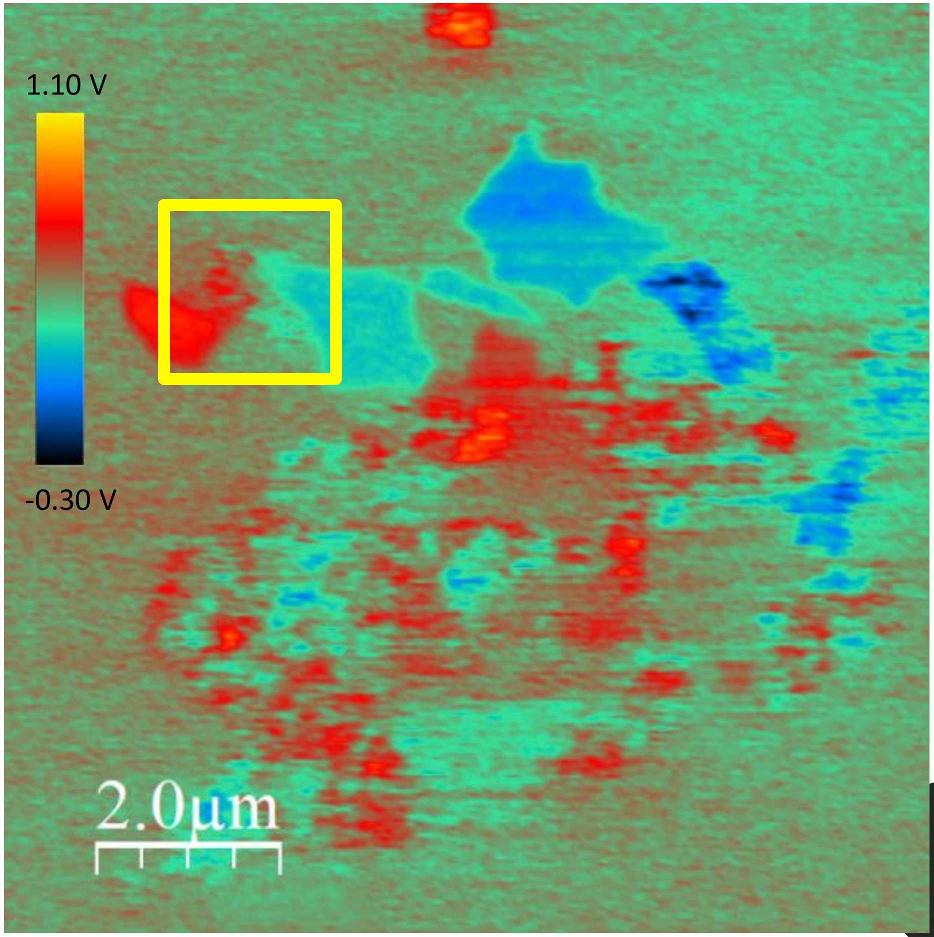

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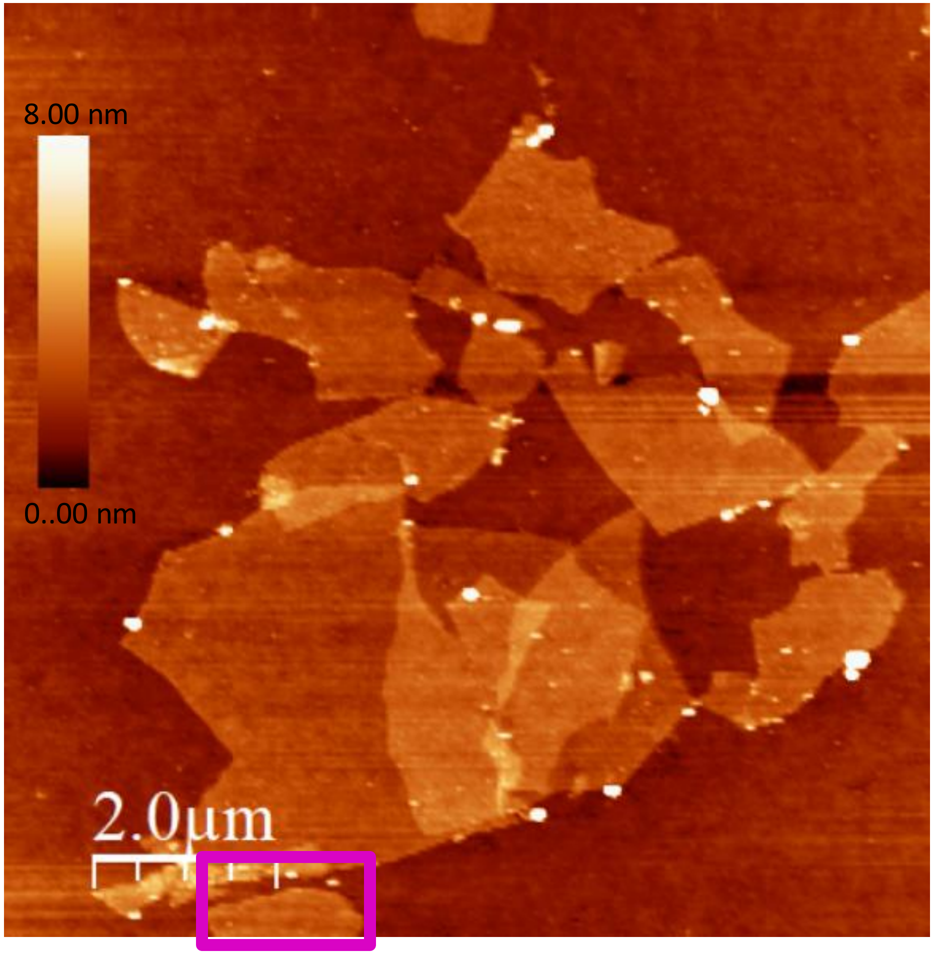

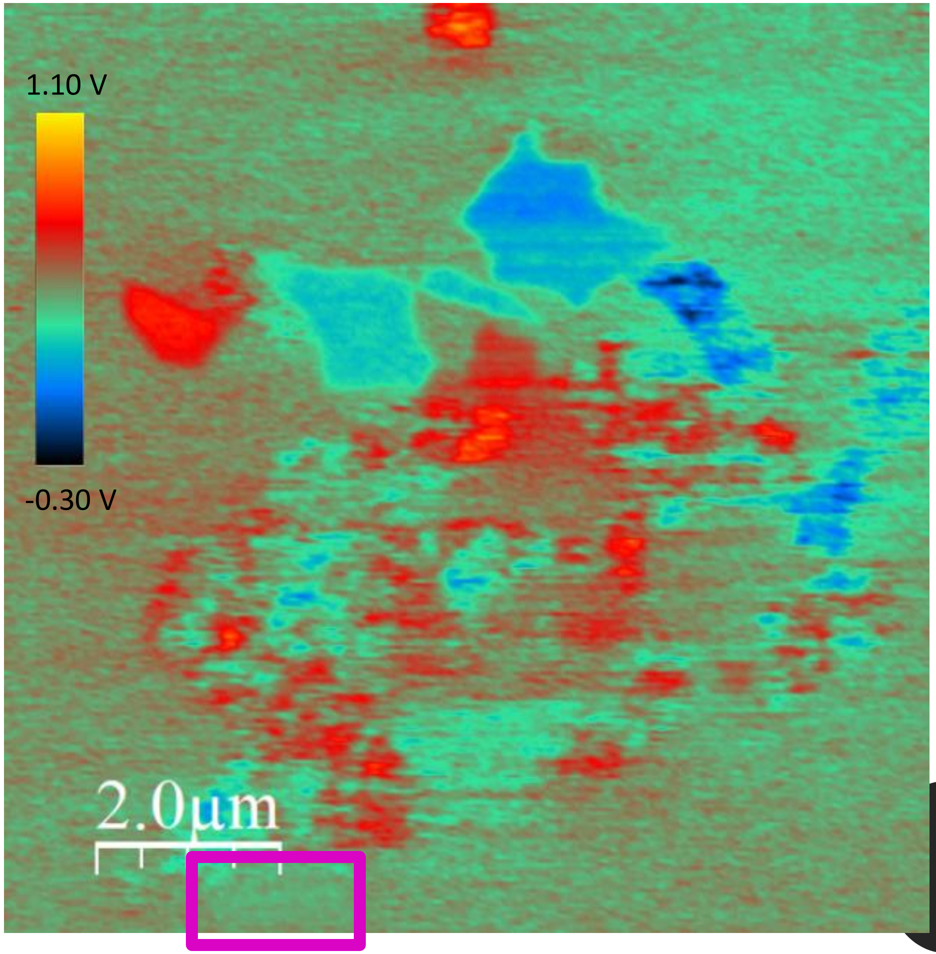

Conclusions

To conclude, we have shown that AFM with KPFM is an ideal technique for studying the electronic properties of disordered materials at the nanoscale. In particular, it has been shown the existence of localized charge domains in GO, due to its disordered nature and randomly arranged functional groups. But also, we have been able to probe the conducting behavior of GO and rGO with charge injection experiments, demonstrating that charge dynamics in rGO are much faster than in GO. Thus, confirming the enhanced conductivity of rGO. The study of these properties is important because charge transport processes occur at these scales.

For future investigations, the intermediate states of reduction could be studied, as their properties can be tuned depending on the degree of reduction. Such versatility opens up avenues for customizing these materials to meet the requirements of several applications.