Synthesis and Properties of Monolayer Graphene Oxyfluoride

Matteo Bruna, B. Massessi, C. Cassiago
A.Battiato, E. Vittone and S. Borini

INRIM (Istituto Nazionale di Ricerca Metrologica)
&
Politecnico di Torino(Italy)

- Overview

Graphene, exceptional material for fundamental research

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Graphene's properties tuning and control

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Graphene's properties tuning and control

applications

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Graphene's properties tuning and control

applications

Chemical route

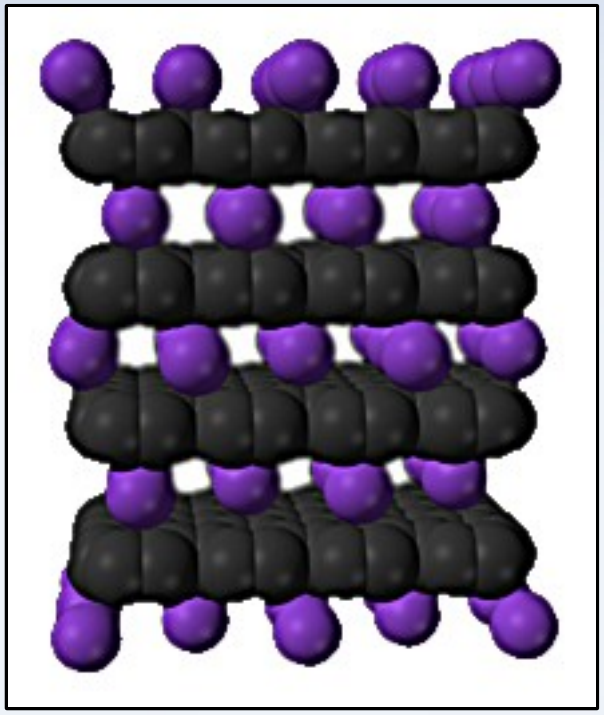
Band-gap engineering

Photonics

Selectivity for sensing applications

Study of new 2D materials

- Graphite intercalation compounds



Courtesy of *wikimedia*

Graphite intercalation compounds

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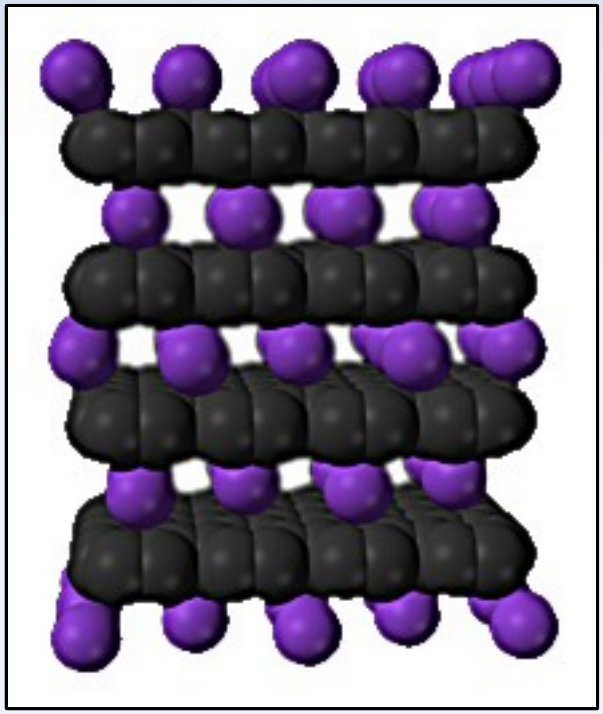
Putting various chemical species between graphene layers in bulk graphite

Chemical bonds can form between carbon atoms and intercalant species

• Graphite intercalation compounds



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Graphite intercalation compounds

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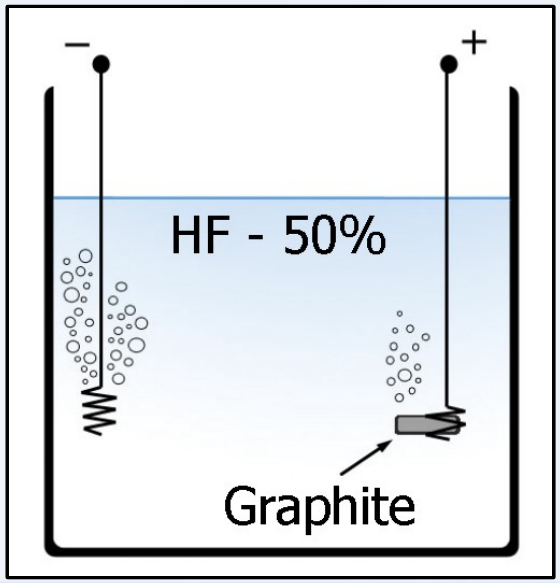
Putting various chemical species between graphene layers in bulk graphite

Simple electrochemical procedure used to intercalate fluorine atoms in graphite



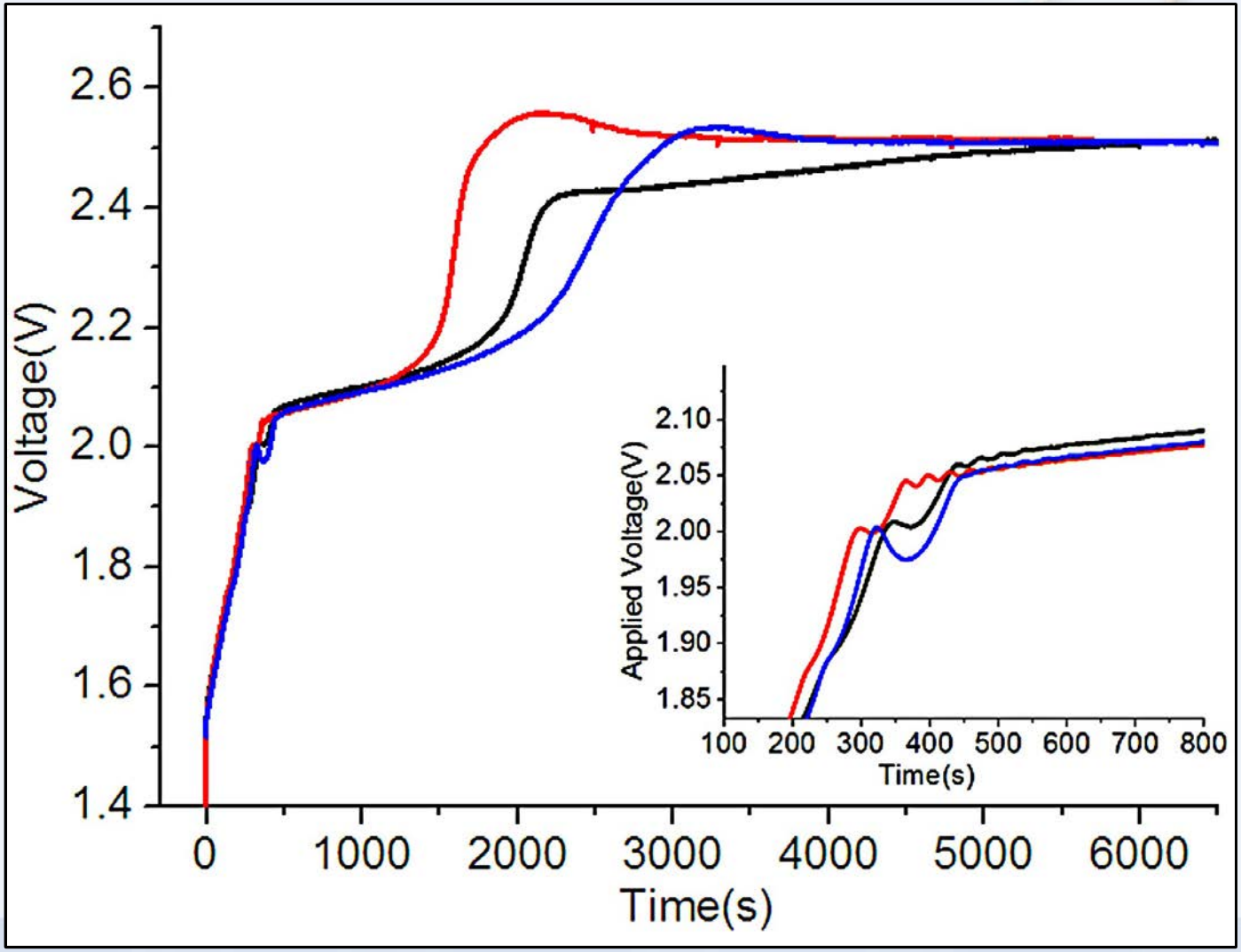
Chemical bonds can form between carbon atoms and intercalant species

Chemically modified graphite ←

Y. Matsuo *et al*, Journal of fluorine chemistry, 87,1998

- The intercalation process – gravimetric analysis

Galvanostatic conditions, I=100mA



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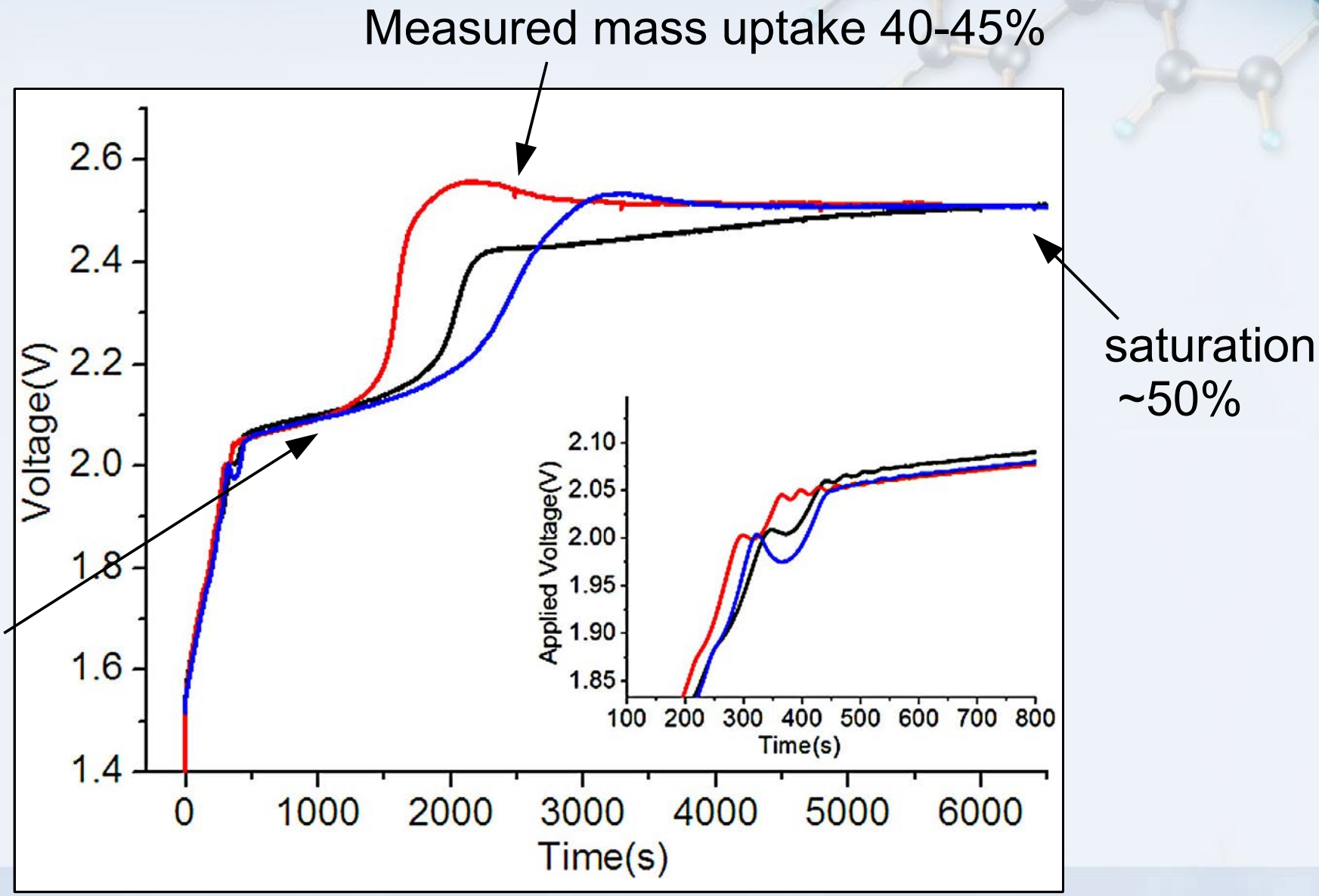
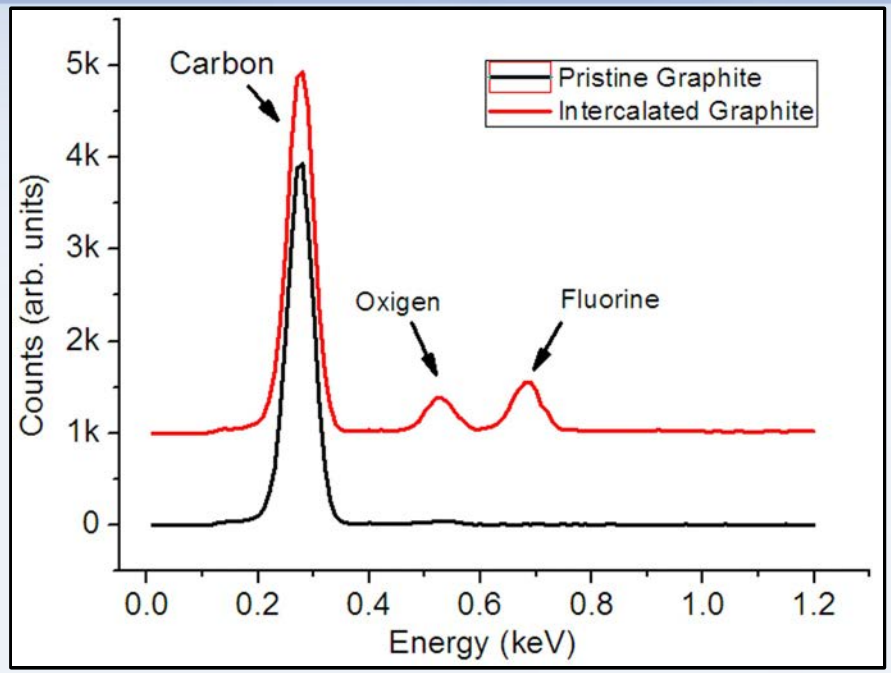


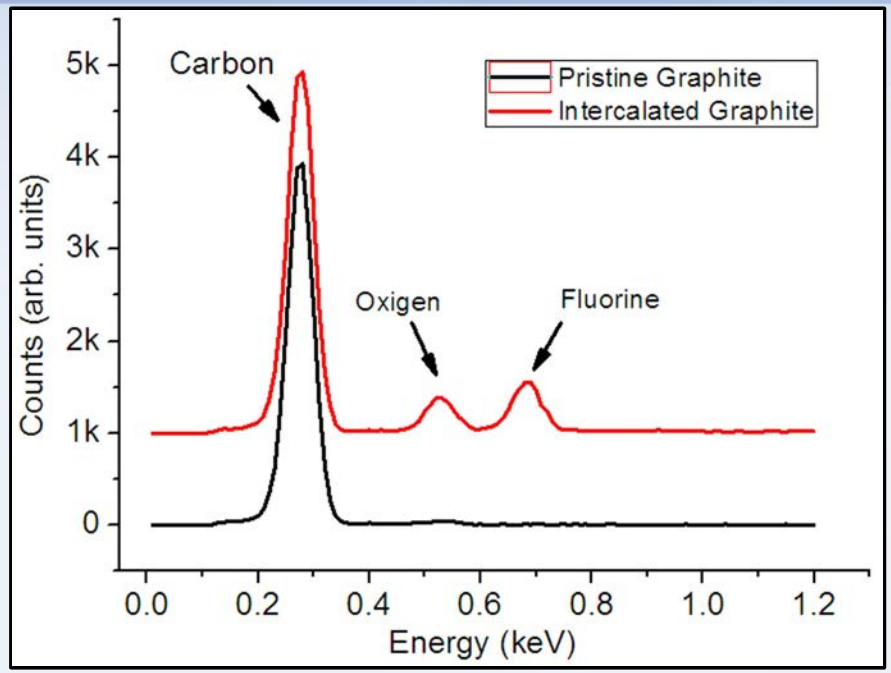
- Chemical analysis



→ Electron Probe Micro-Analysis

presence of: -Carbon
-Fluorine
-Oxygen

- Chemical analysis



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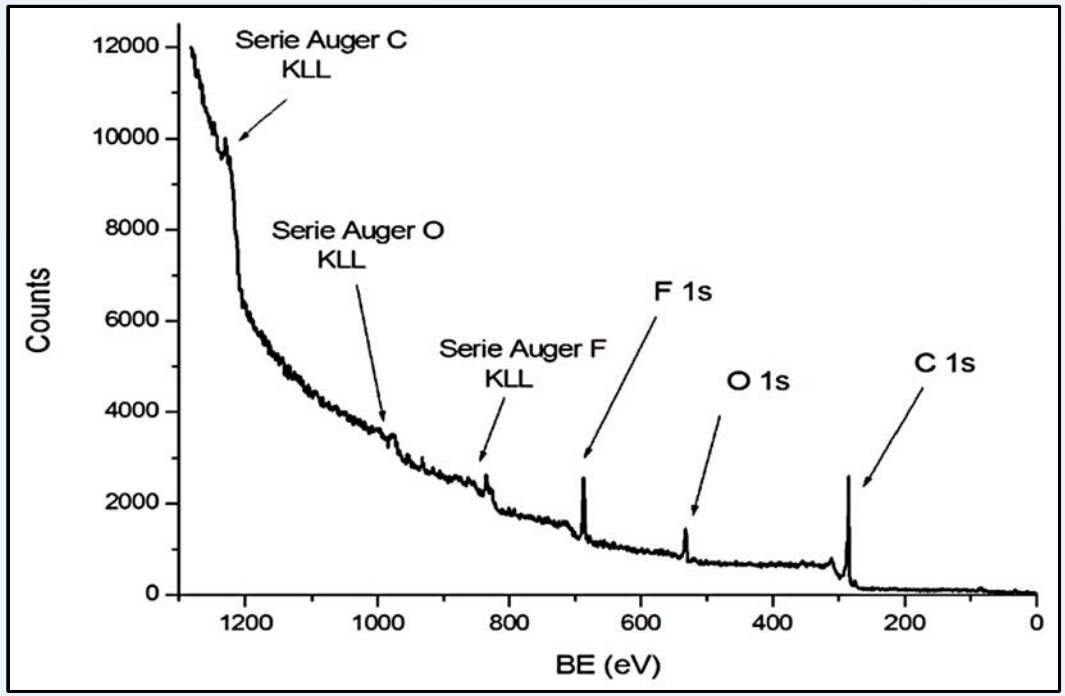
XPS: Presence of various chemical bonds

C-C
C-F covalent and semi-ionic
C=O
C-O-H



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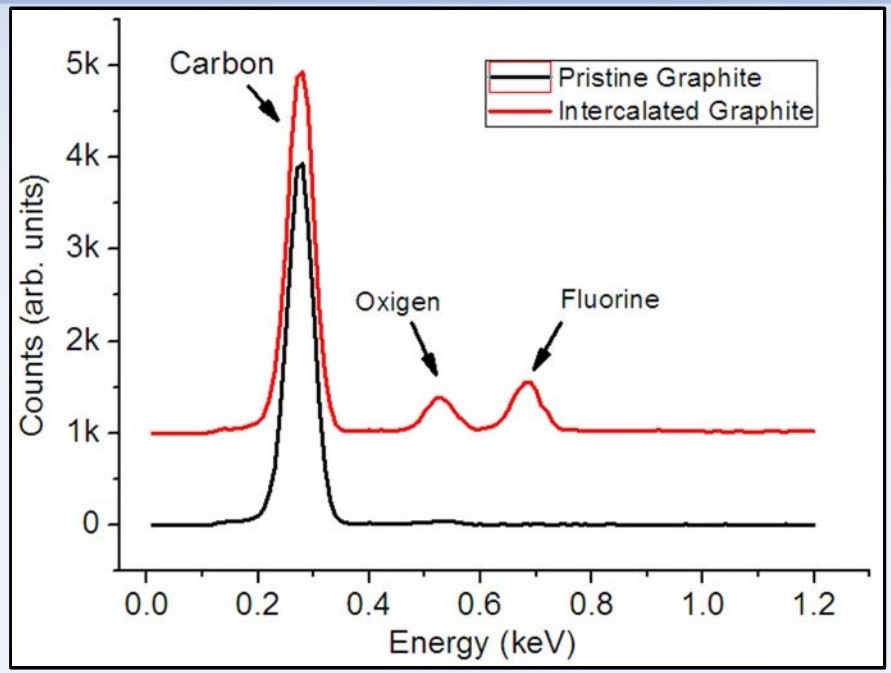

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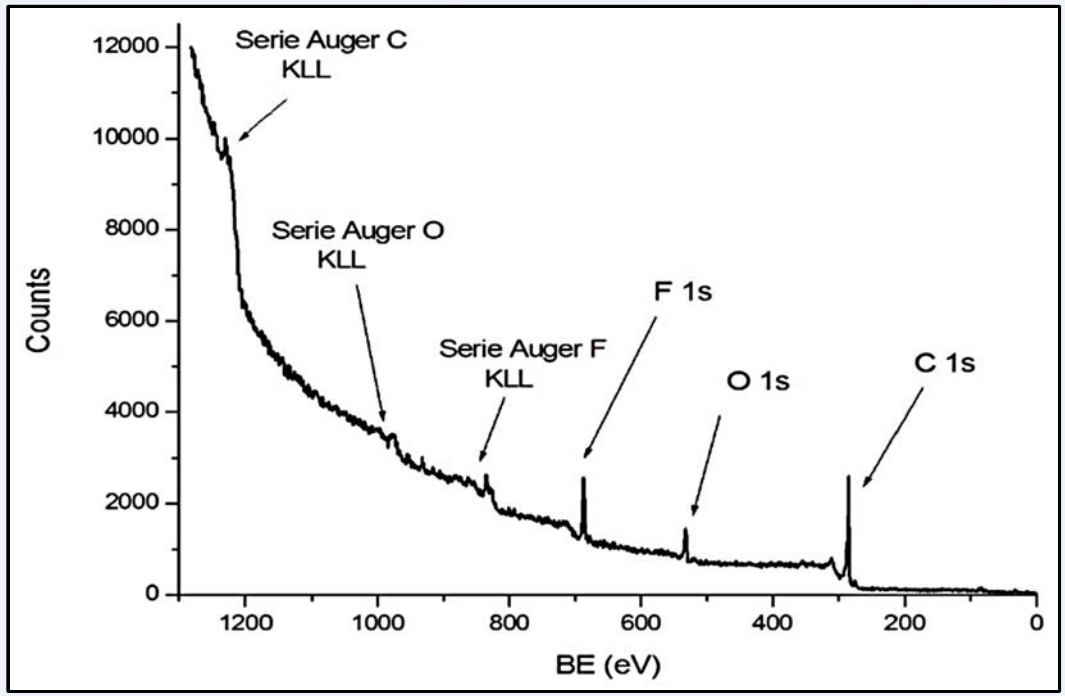

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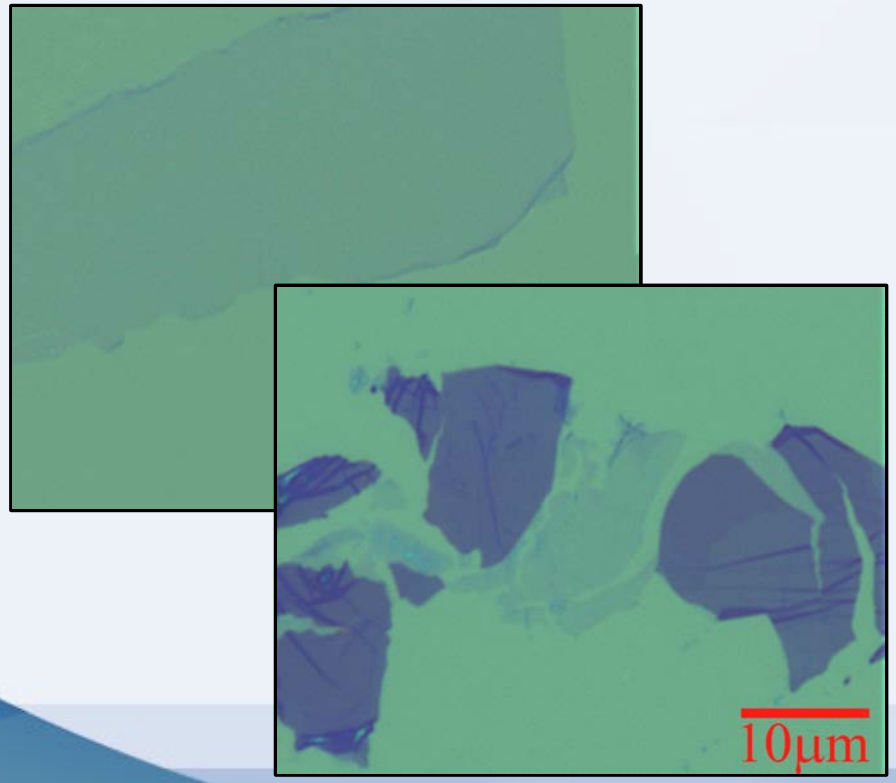
Evidence for covalent modification of graphite

- Tape deposition, Oxyfluorinated thin layers

Tape exfoliation / deposition applied on chemically modified graphite

Large, optically homogeneous flakes of Oxyfluorinated graphene(OFG)



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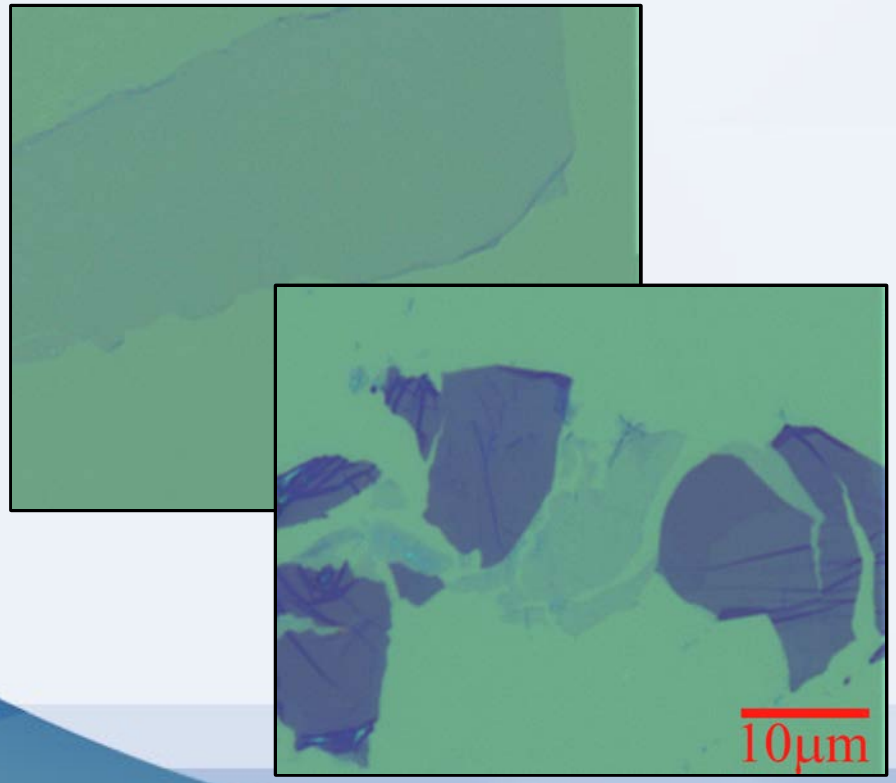
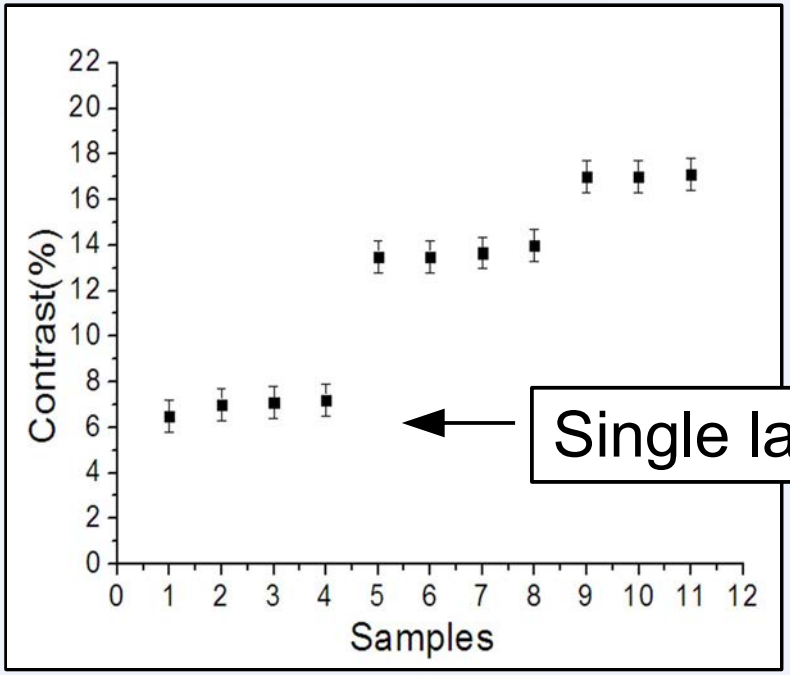
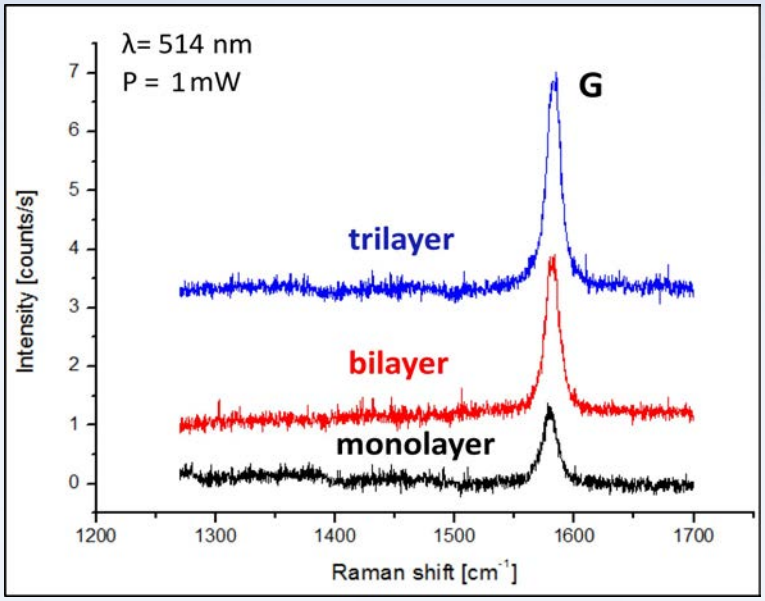


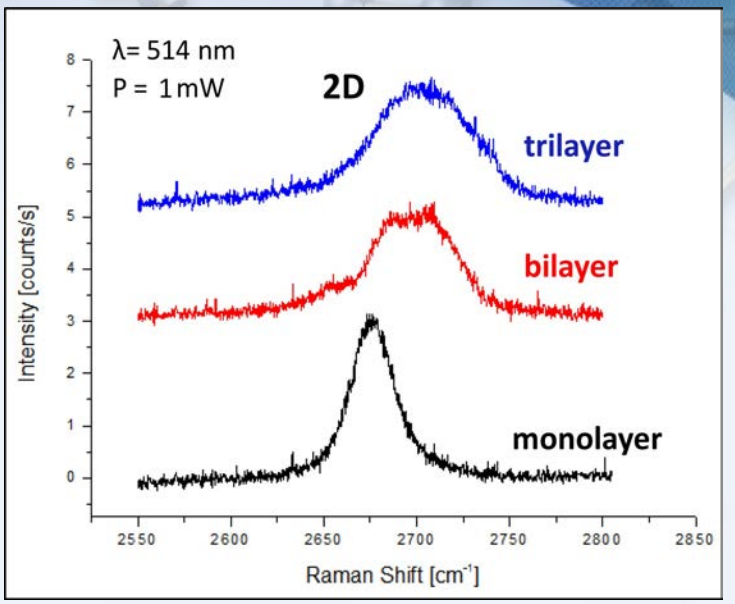


Single layer(???)

• Raman Spectra of Oxyfluorinated graphene

Pristine Graphene by HOPG exfoliation →





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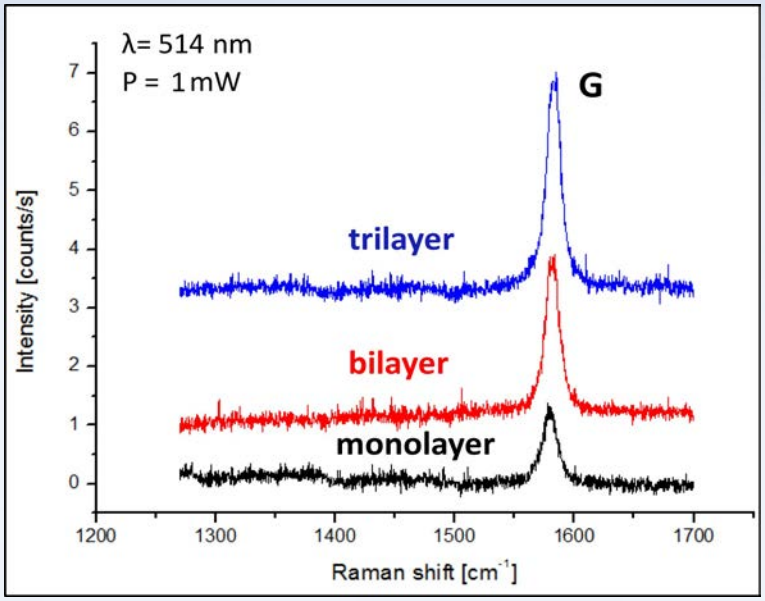
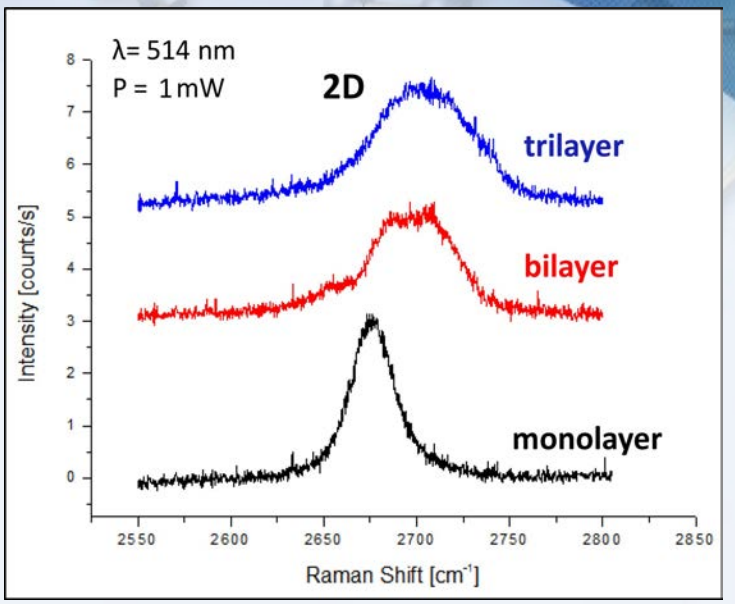
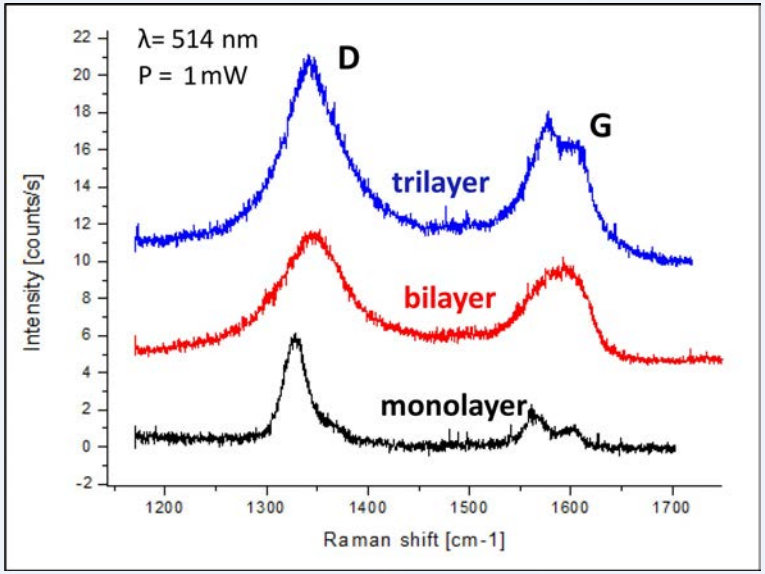
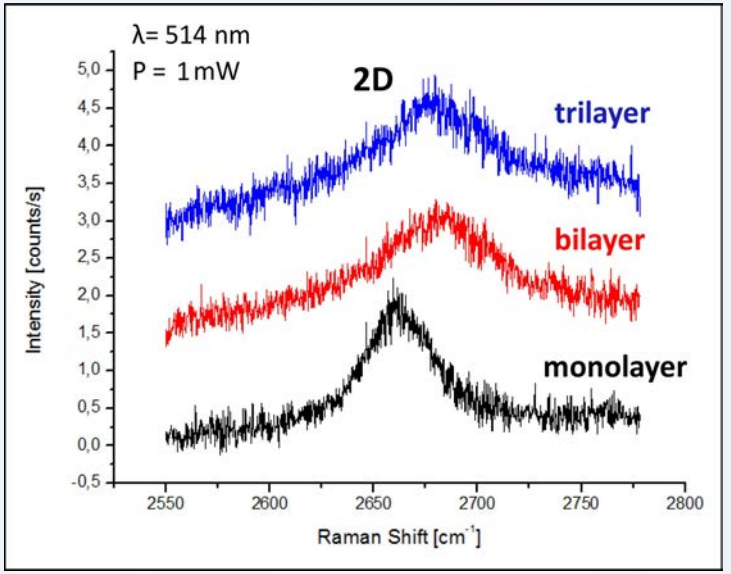


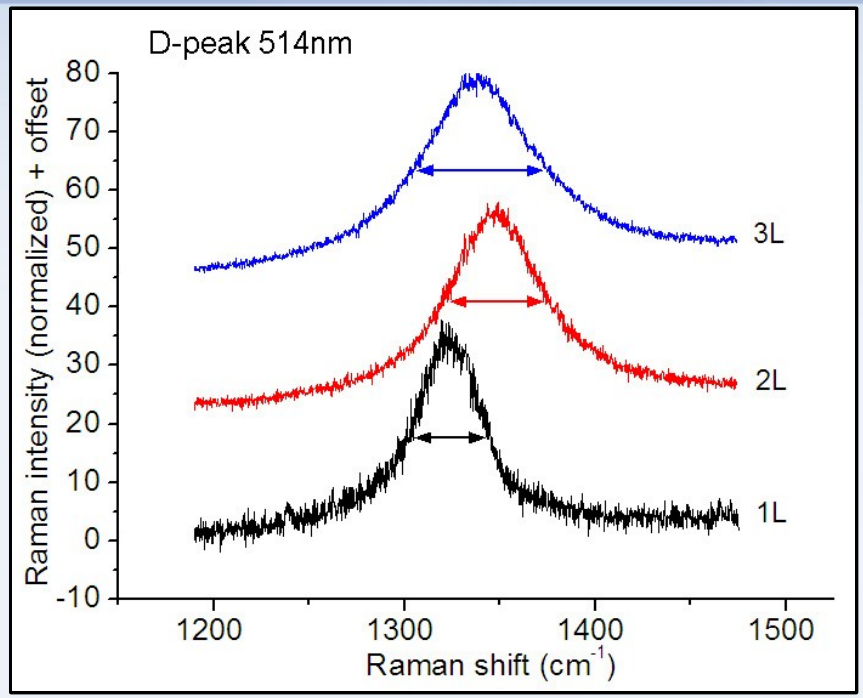


Oxyfluorinated graphene by intercalated graphite exfoliation →





• Single OFG layer identification



D peak FWHM increases with number of layers

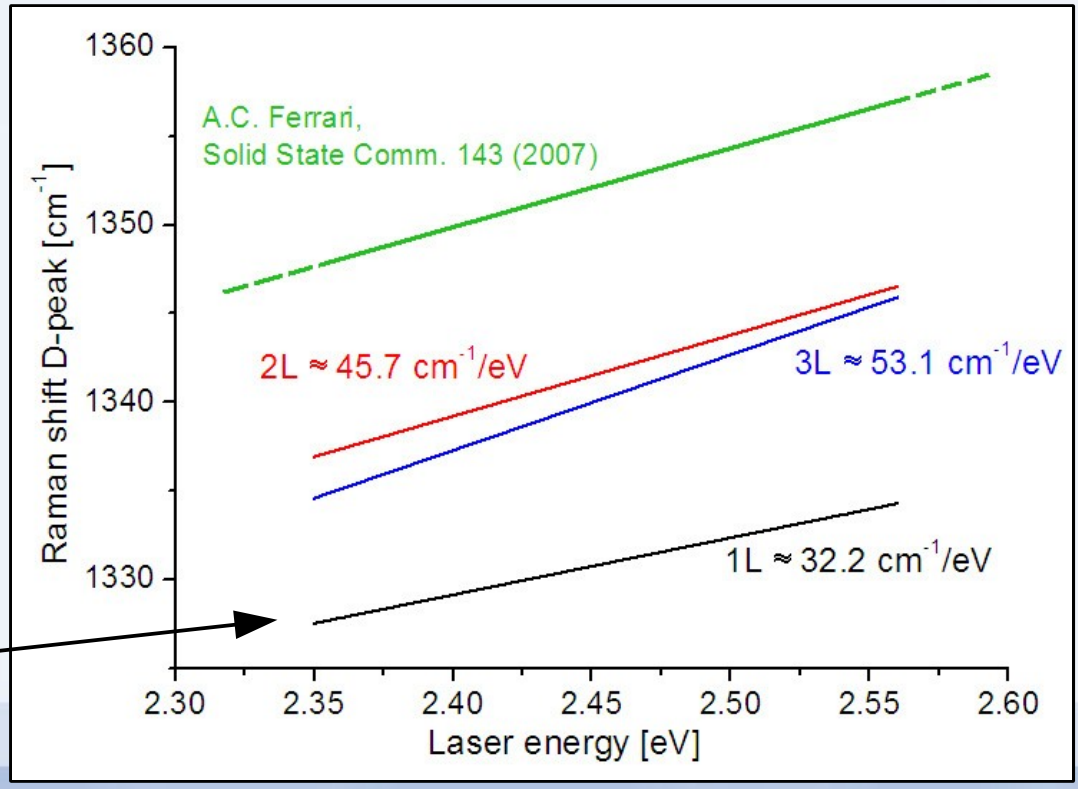
	FWHM D peak [cm^{-1}]
monolayer	28,8 ± 6,4
bilayer	68,0 ± 12,3
trilayer	84,7 ± 10,4

Analysis made on more than 150 OFG flakes

Phonon dispersion with laser frequency still observable →

Phonon energy reduced by fluorine [Gupta et al. Journal of fluorine chemistry, **110**, 145-151(2001)]

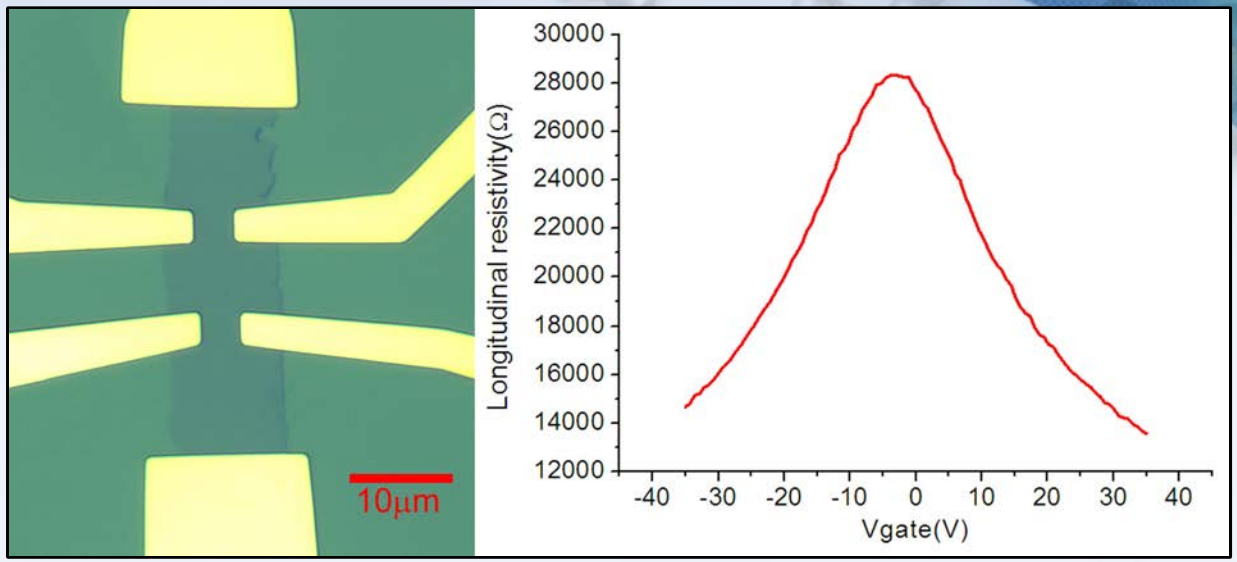
Higher reduction for monolayers



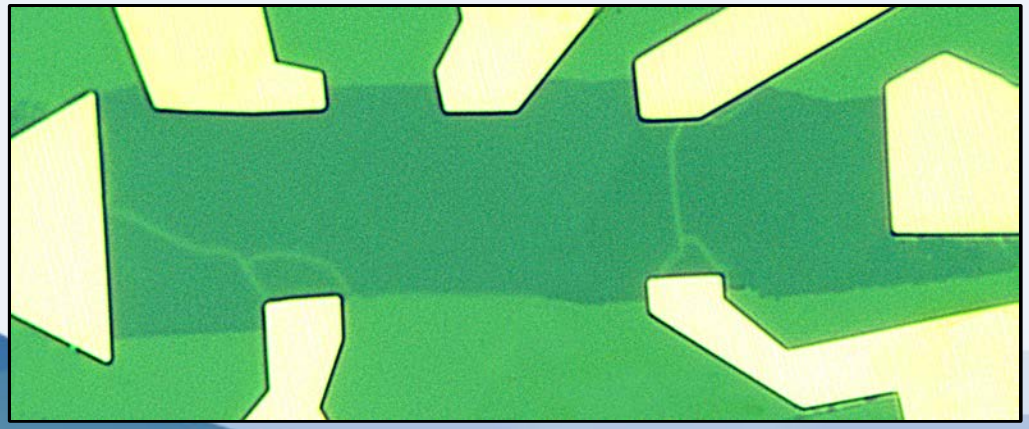
• Device fabrication

Electron-Beam Lithography
+
Metal evaporation
+
Lift-off

(10nm Ti + 50 nm Au) ⟶



Graphene mechanical properties reduced upon chemical modification



Graphene-like ambipolar field effect
Tuning of charge carriers density by back-gate

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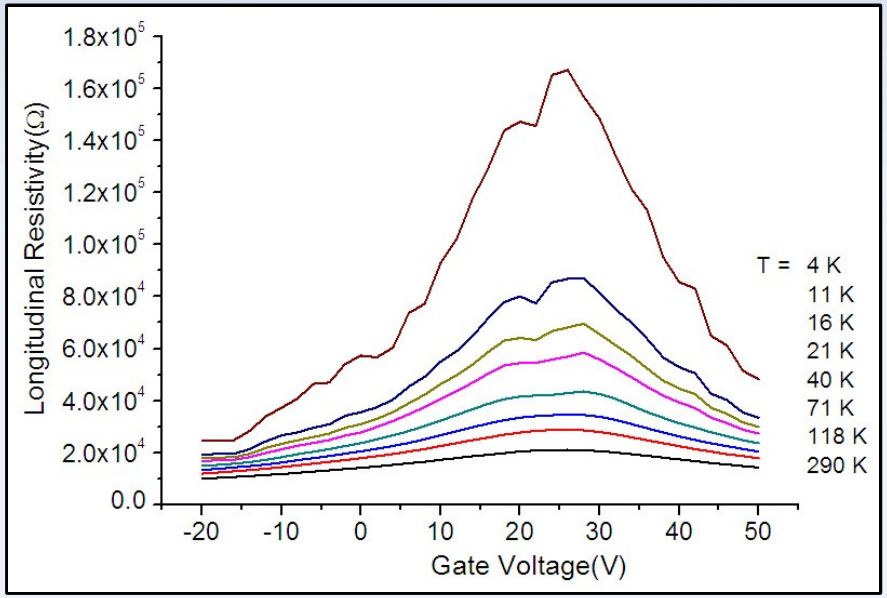
Room temperature mobility

~150$cm^2V^{-1}s^{-1}$

• Low-T transport measurement

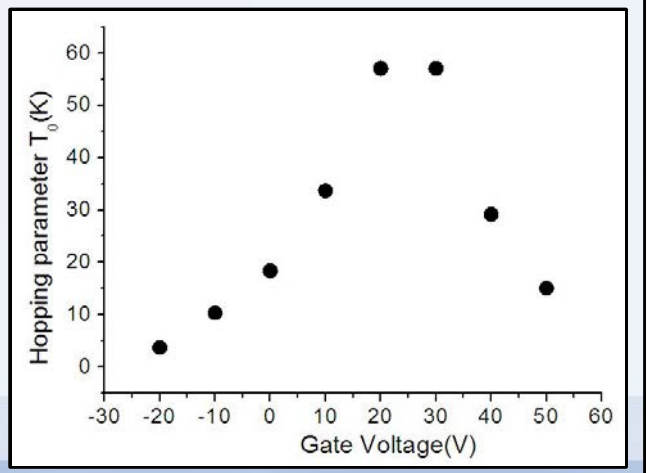


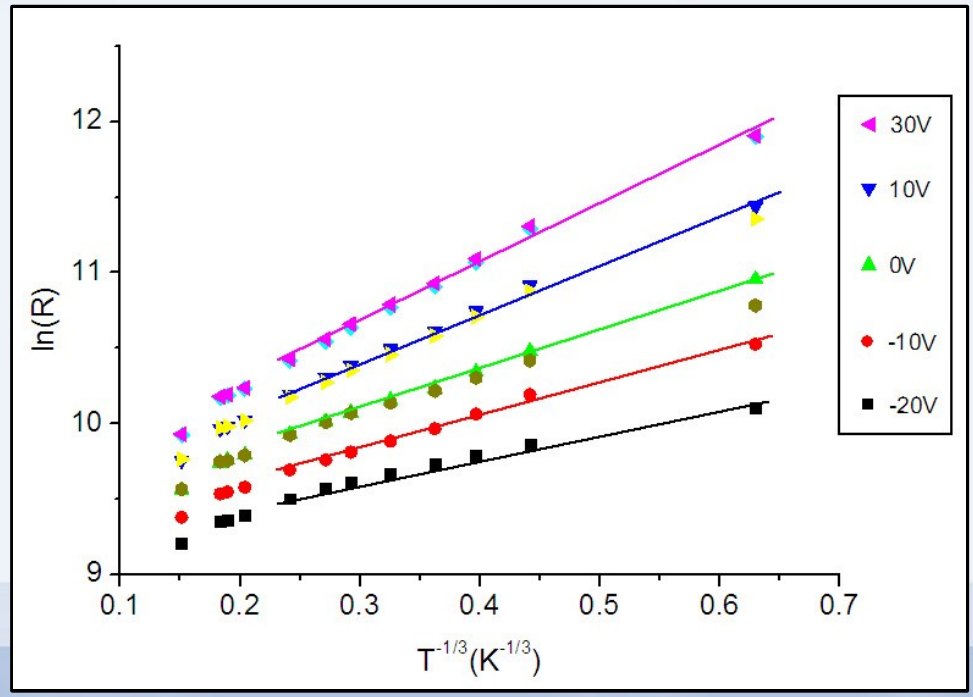
Resistance near the charge neutrality point increases by one order of magnitude going from 290 to 4K

Temperature behaviour well described by the 2D Mott variable-range hopping model

$$R(T) = R_0 \, EXP[T_0 / T]^{1/3}$$

Doping-dependent hopping parameter T_0

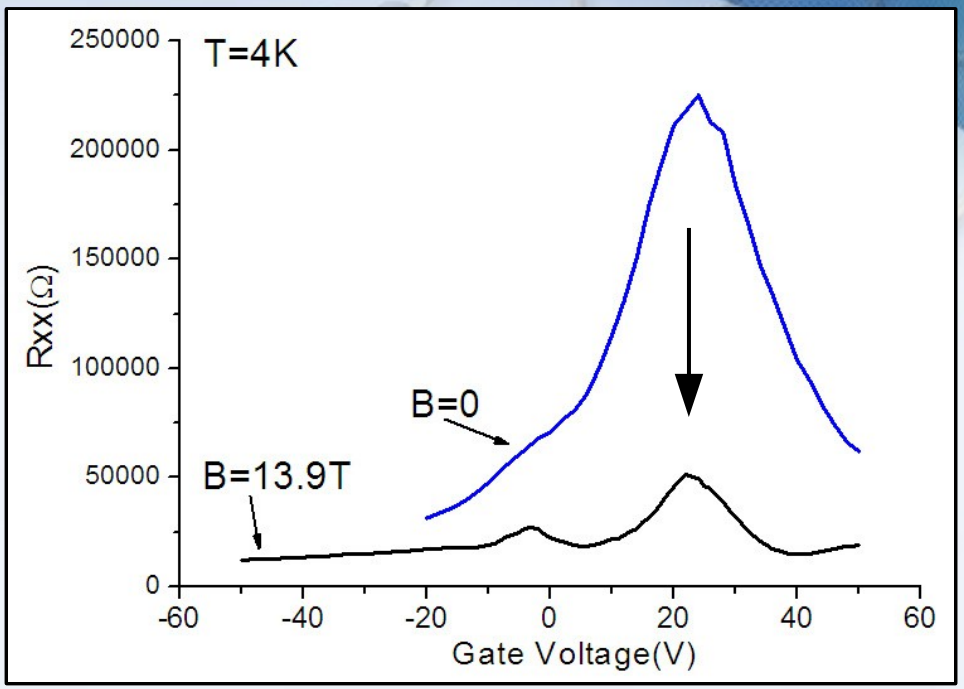


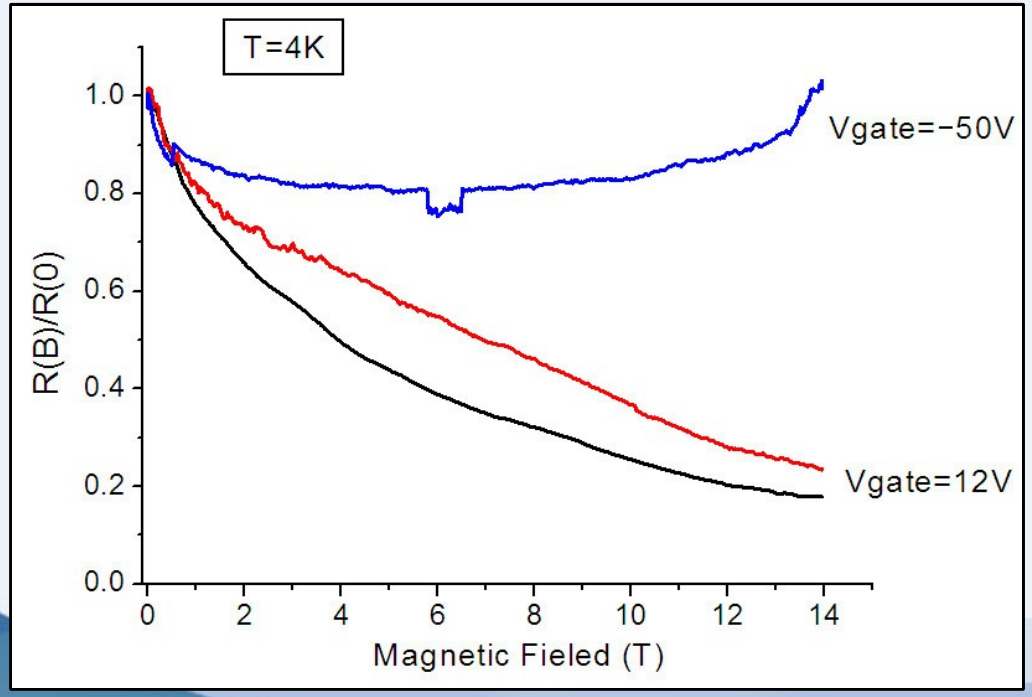


- B-field transport measurements

Huge magnetoresistance

Weak signatures of Shubnikov–de-Haas oscillations





High carrier density → Weak localization

Dirac Point → Strong localization

[Hong *et al*. PRB 83, 085410 (2011)]

• Summary

•Very easy method for covalent modification of graphite

•Successful exfoliation of thin layers

•Contrast/Raman signature of Oxyfluorinated graphene monolayers

Material interesting for:

•Subsequent chemical functionalization

• Transport study in disordered 2D systems

•Possibility of performing different intercalations

→ Chemical engineering of 2D semiconductors