Graphene Canada

Charge Transport in Polycrystalline Graphene-Based Materials

Stephan Roche

stephan.roche@icn.cat

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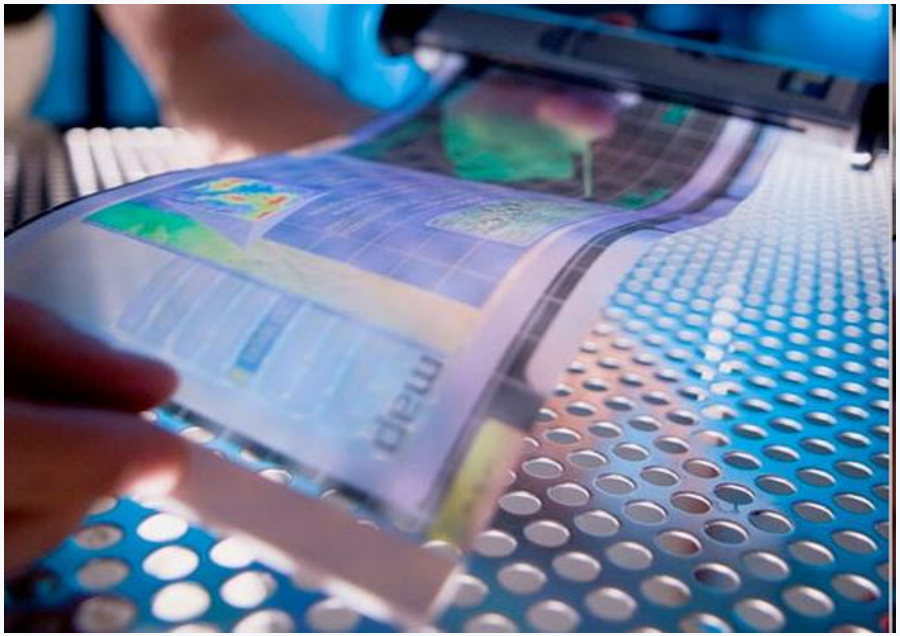
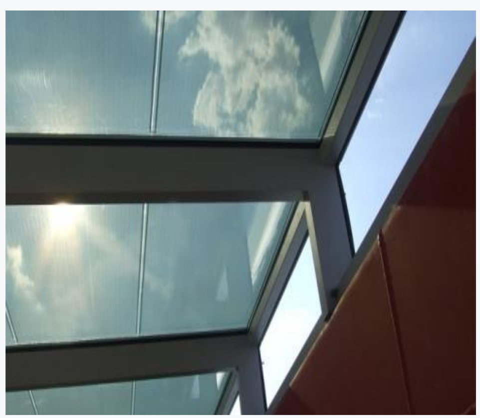
EXCELENCIA SEVERO OCHOA

What type of “graphene material” is relevant for applications ?



Trade off between structual quality
Large scale mass production

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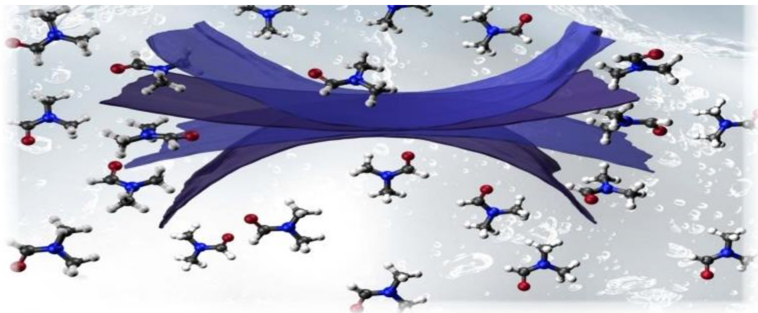
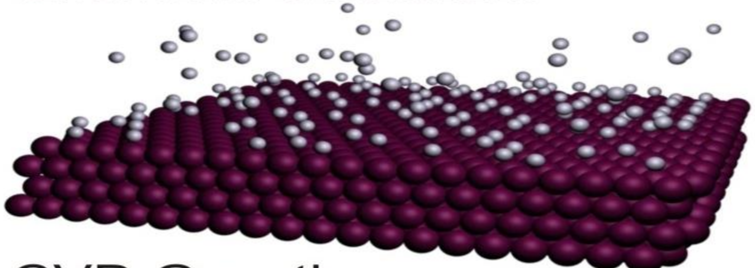
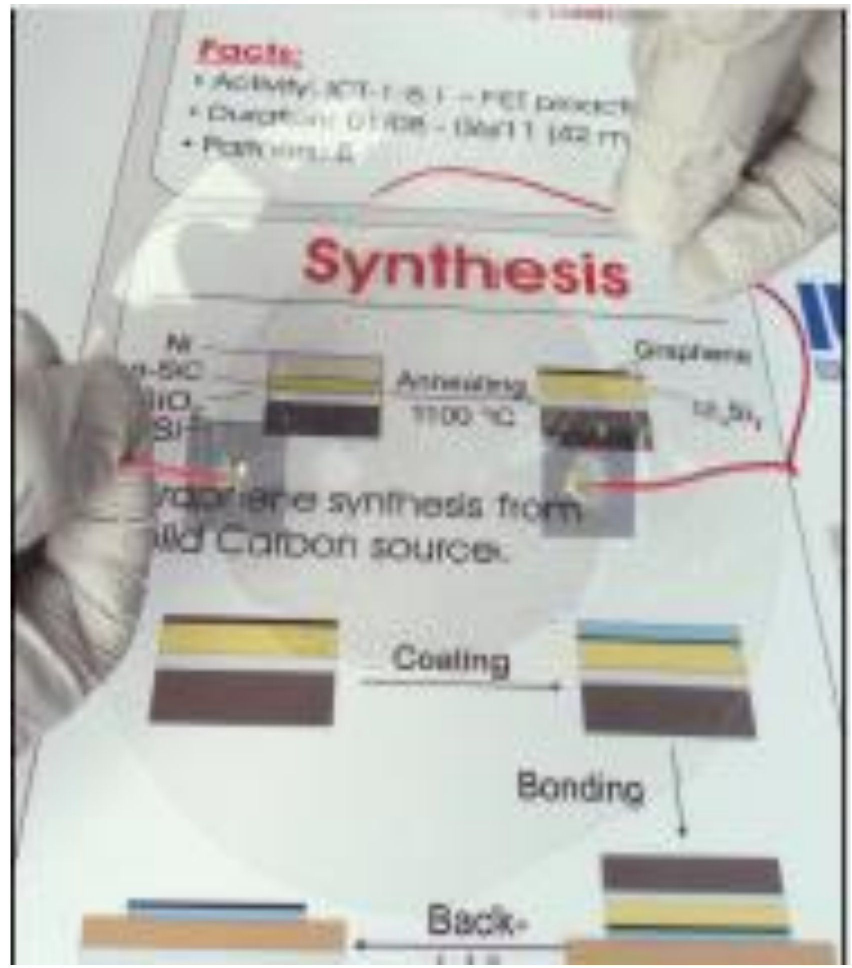


Chemical exfoliation



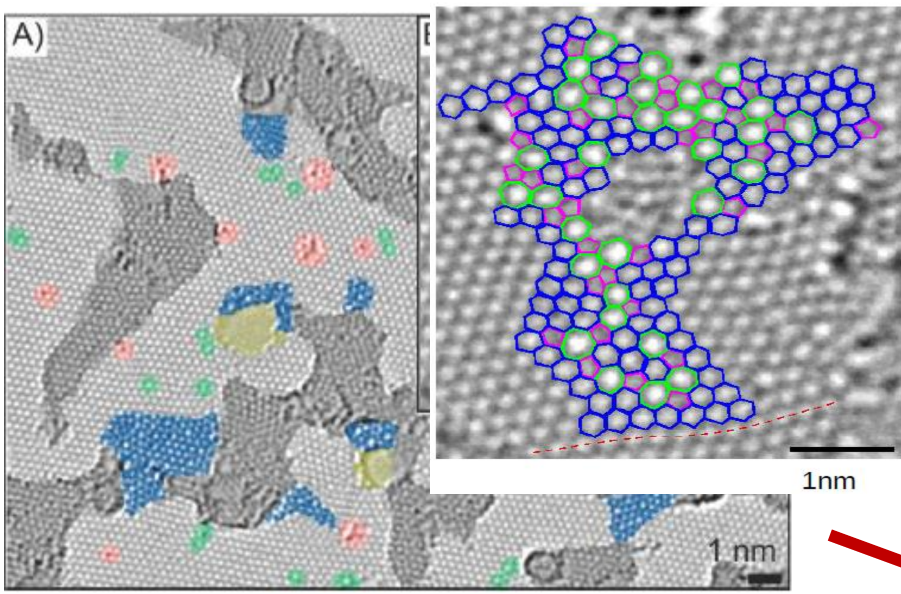
CVD Growth



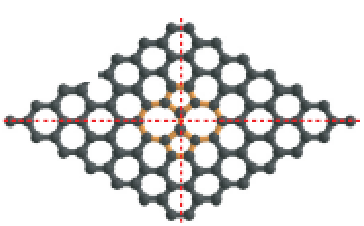
AMO

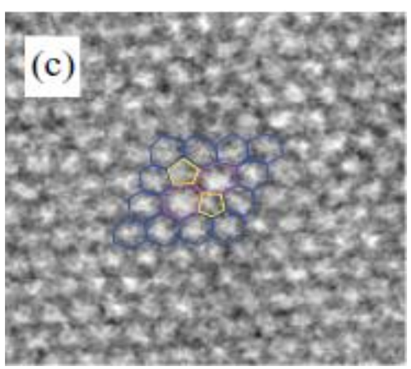
Reduced graphene oxides (rGO)

Dozens of methods based on chemical, thermal or electrochemical means



Stone-Wales



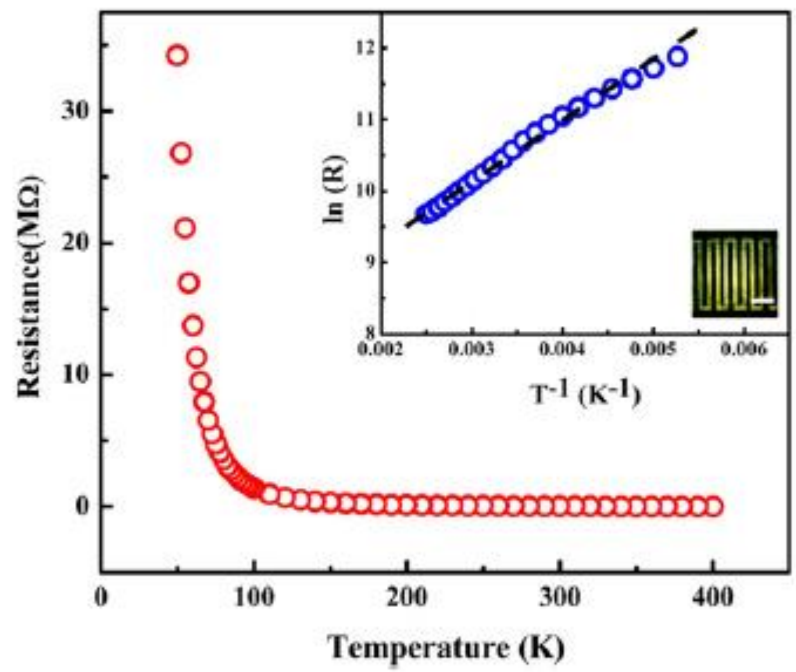


Gomez-Navarro et al **Nano Lett. (2010)**

The obtained rGO exhibit high density of structural defects

- pentagons/heptagons, octogons,…

Very large zoology from single defects to large areas

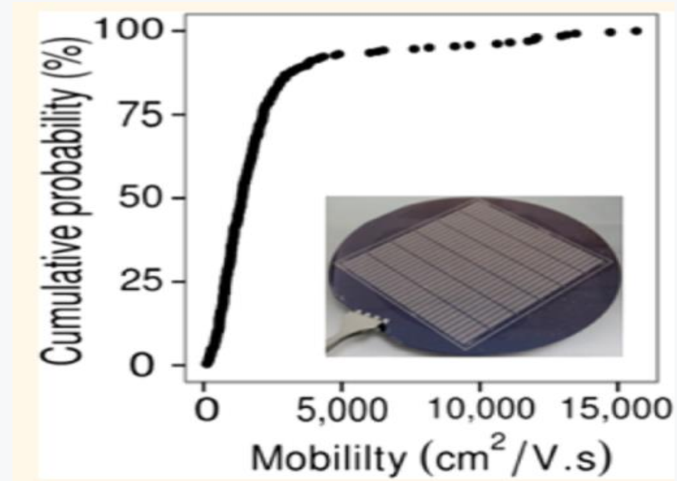


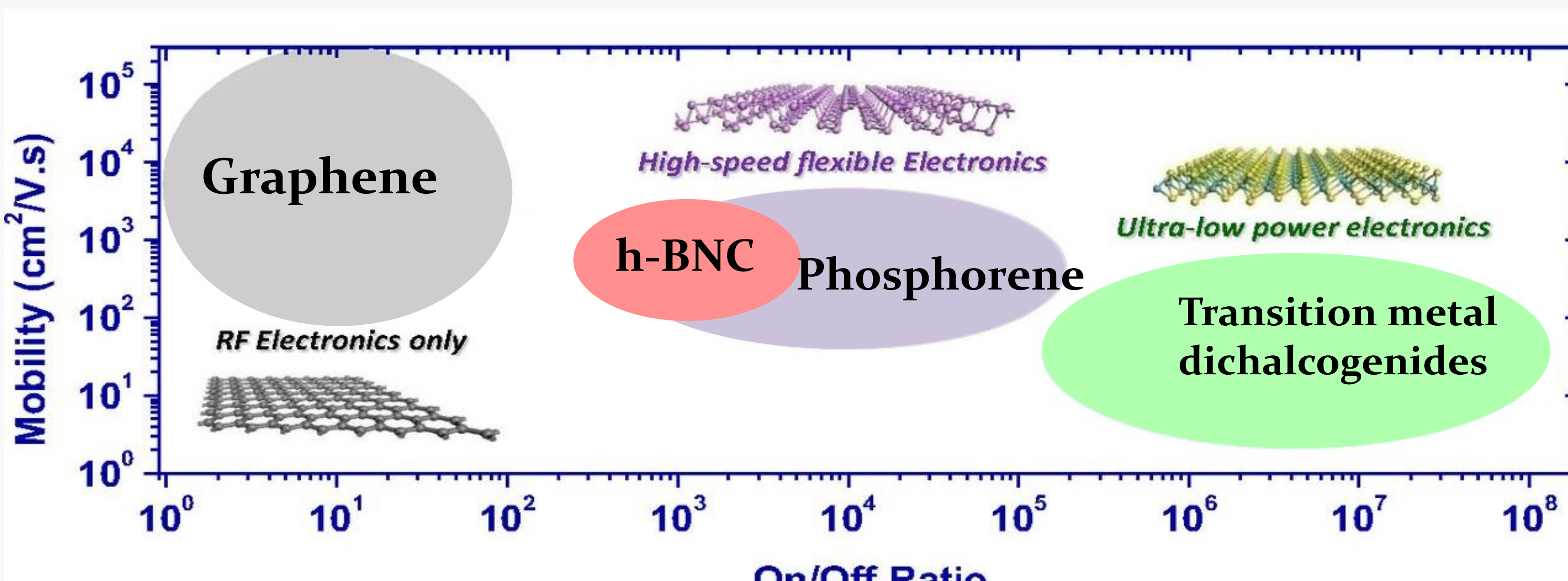
Muchharla et al **2DMaterials 1, 011008 (2014)**

300 mm wafer-scalable 2D Materials

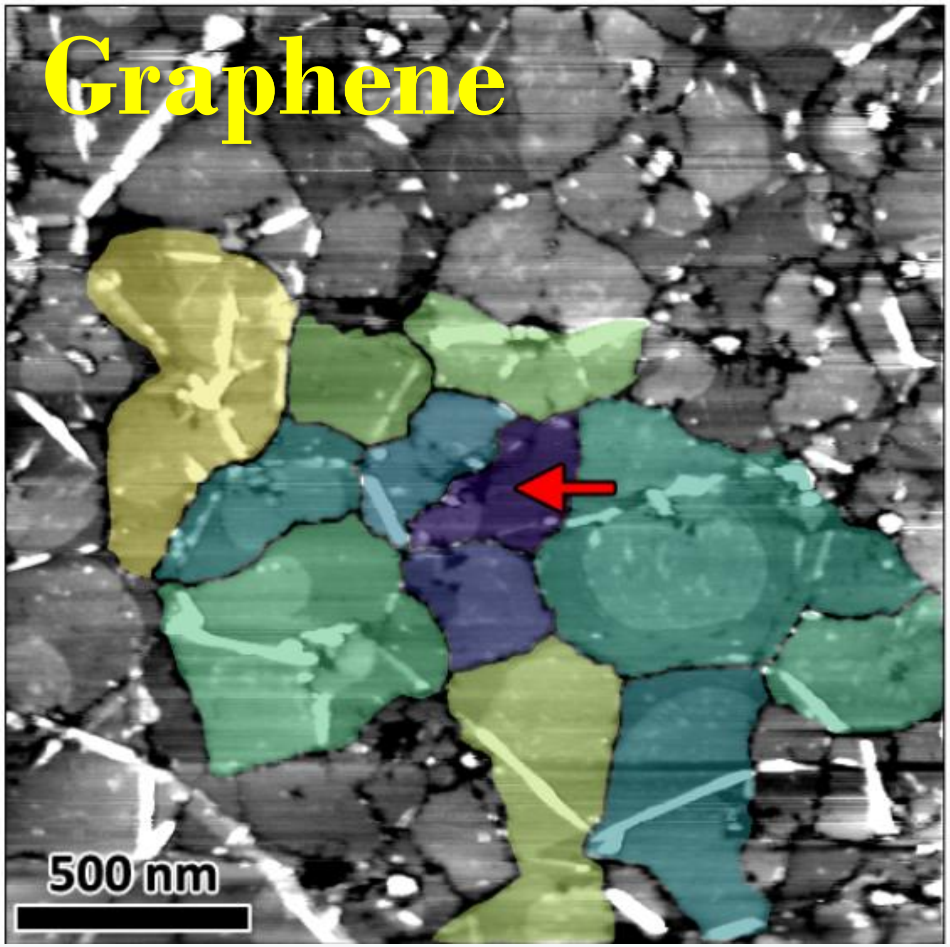
AIXTRON

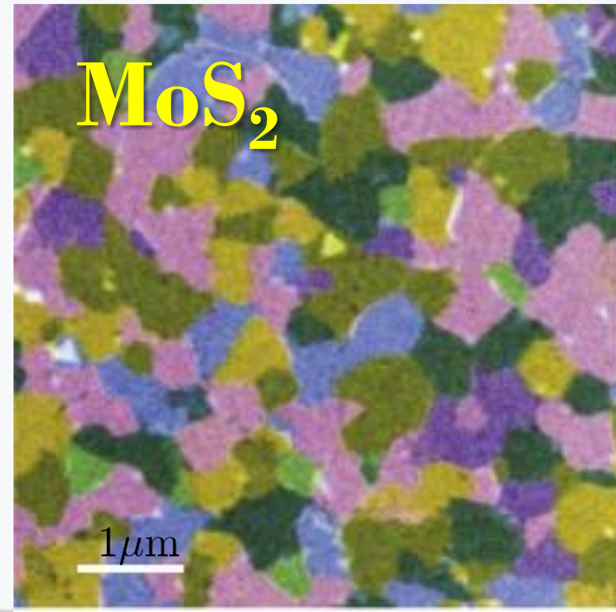
Ken Teo and
Deji Akinwande
ACS Nano **2014, 8 (10), 10471–10479**

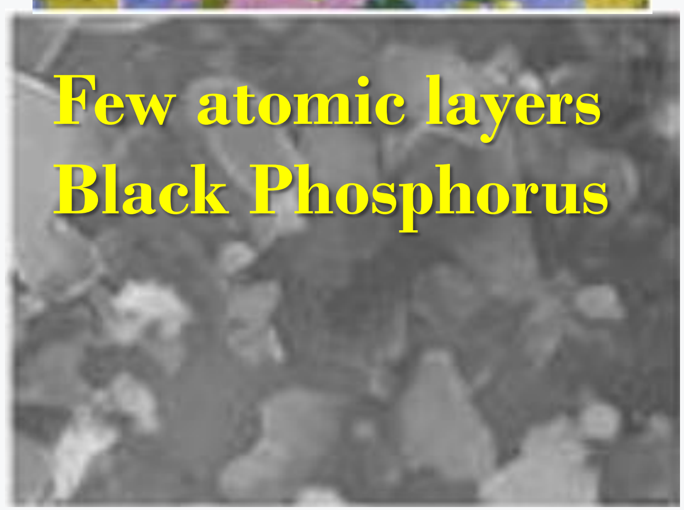




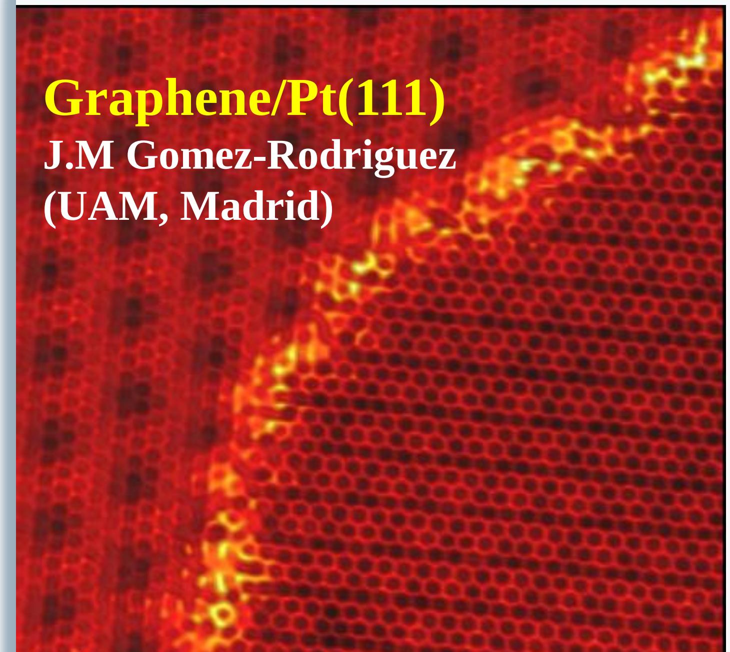
CVD-Grown 2DMaterials
Polycrystalline morphology





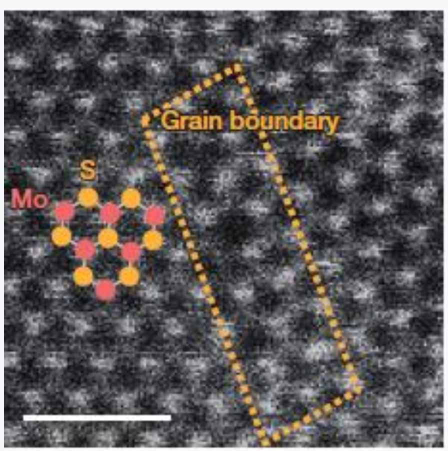


Grain boundaries

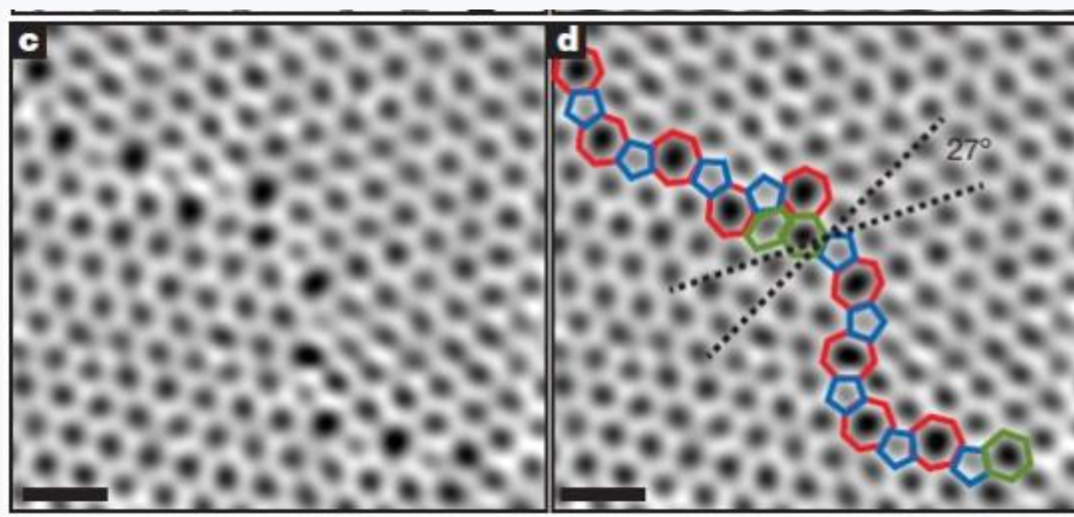


Long range 1D defects (“dislocations”)

1. formed by odd-membered rings
(pentagons, heptagons, heptagons,..)
2. large contamination of the GBs
(chemical reactivity strongly enhanced)



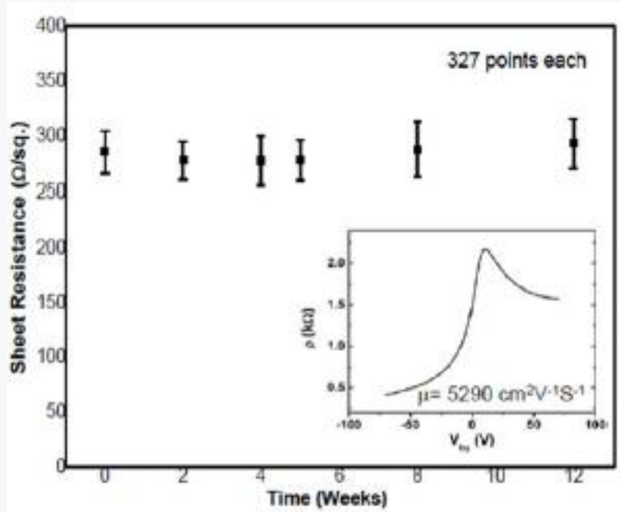
Kang et al, **Nature 2015**

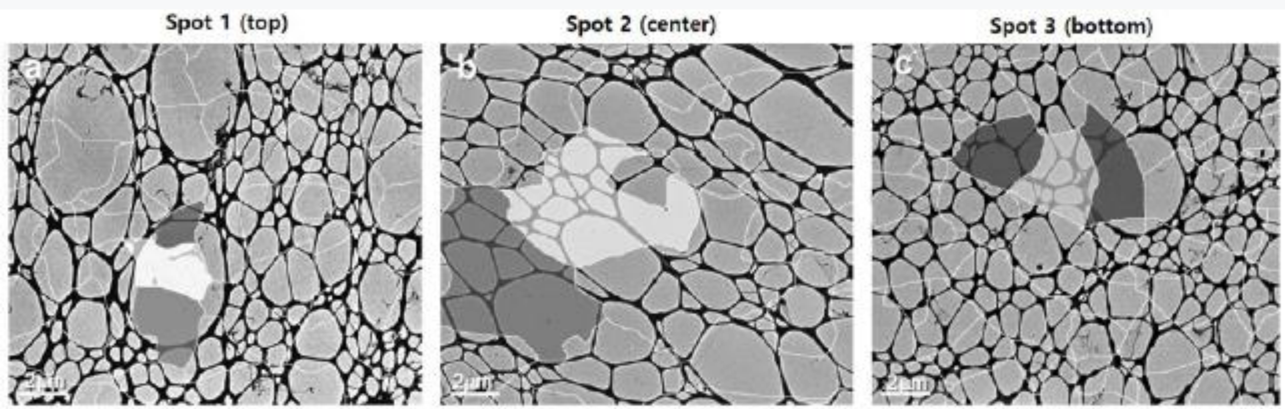


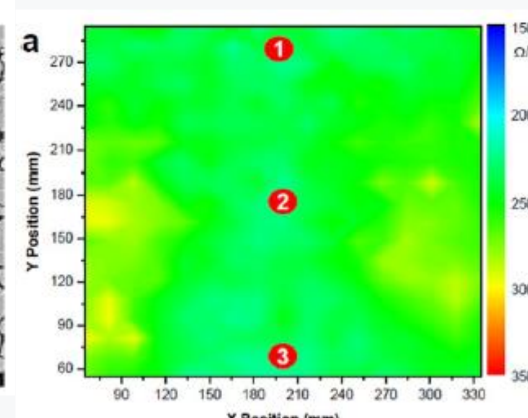
Huang et al, **Nature 2011**

How polycristalline morphology affects device performances?

Rapid Thermal-CVD
Graphene growth rate
1400 cm²/h



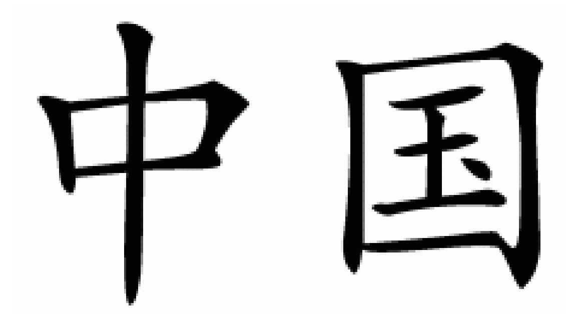




Ryu et al., ACS Nano 8, 950 (2014)

MARCH 2015

Chinese company announces Graphene-based smartphones



The Galapad Settler utilises **graphene in its design to increase battery life and touchscreen sensitivity**

“Graphene is used in in the 5.5-inch phone’s touchscreen, 3000-mAh battery and case, said a representative for Moxi who gave his name as Mr Wu.”

Chongqing-based graphene researcher and maker Moxi teamed up with Shenzhen-based tablet maker Galapad to release 30,000 of the Android handsets this week, according to their websites.
Each device costs 2,499 yuan (**US$399**)

Photo: Galapad

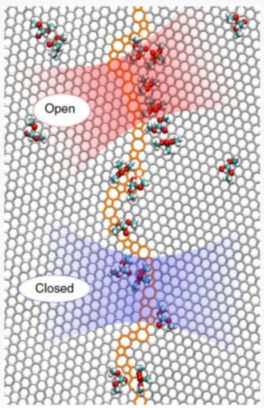
Graphene Grain boundary-based sensors

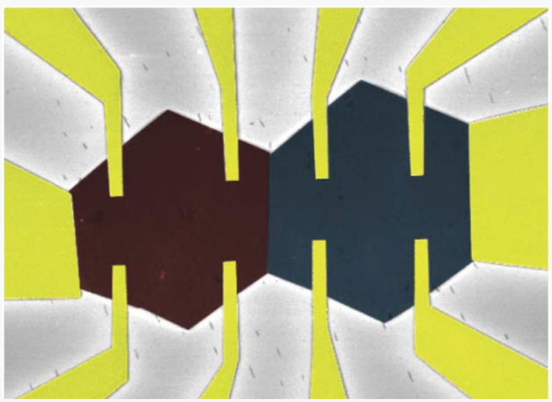
2014. U.S. patent application No. DH073, filed June 2014

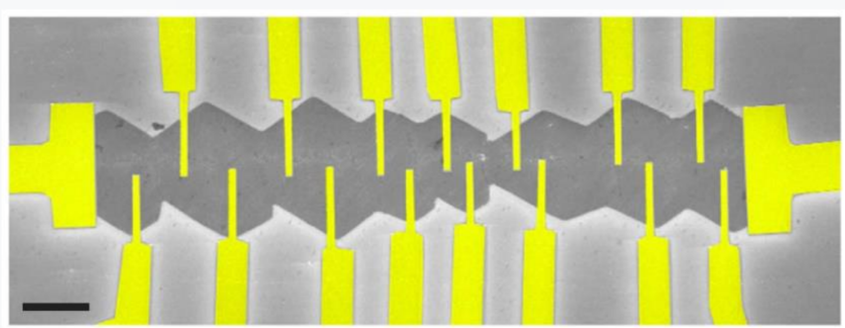
50 ppb. of DMMP Dimethyl methylphosphonate

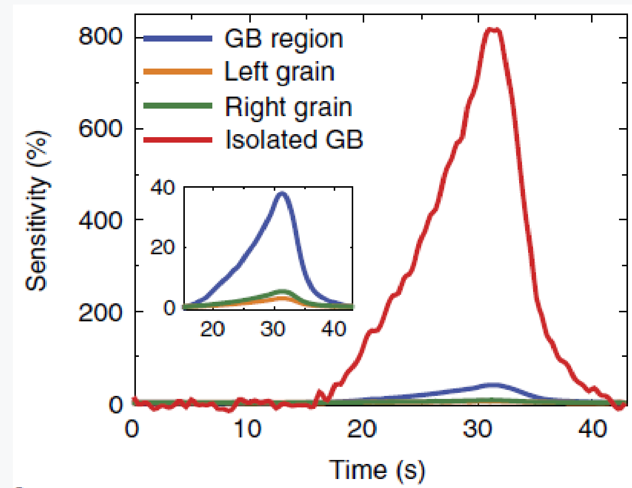
300% higher

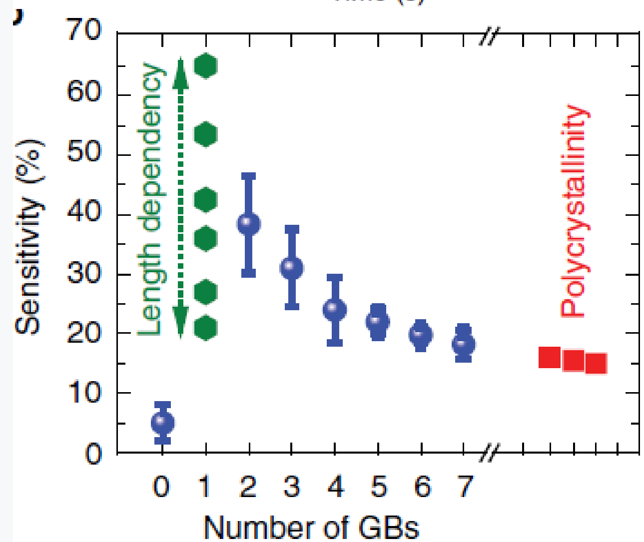
Sensitivity to gas exposure $= \frac{R_{\text{after}} - R_{\text{before}}}{R_{\text{before}}}$











Salehi-Khojin *et al.*,

Nature Communications 5 4911 (2014)

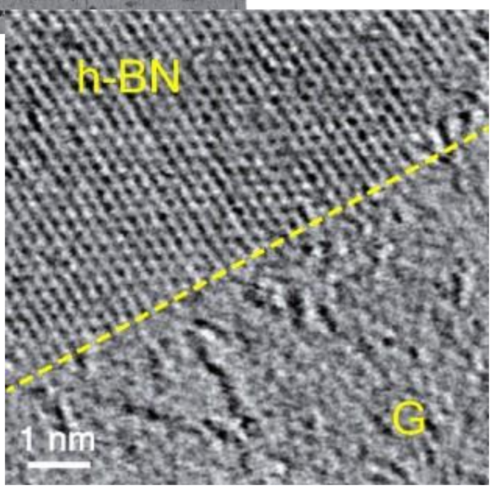
Hybrid graphene/h-BN atomic layers

Precise 2D domains of graphene and h-BN are stitched together,

Combined electronic properties

Create periodic arrangements of domains with

size ranging from tens of nanometres to millimetres

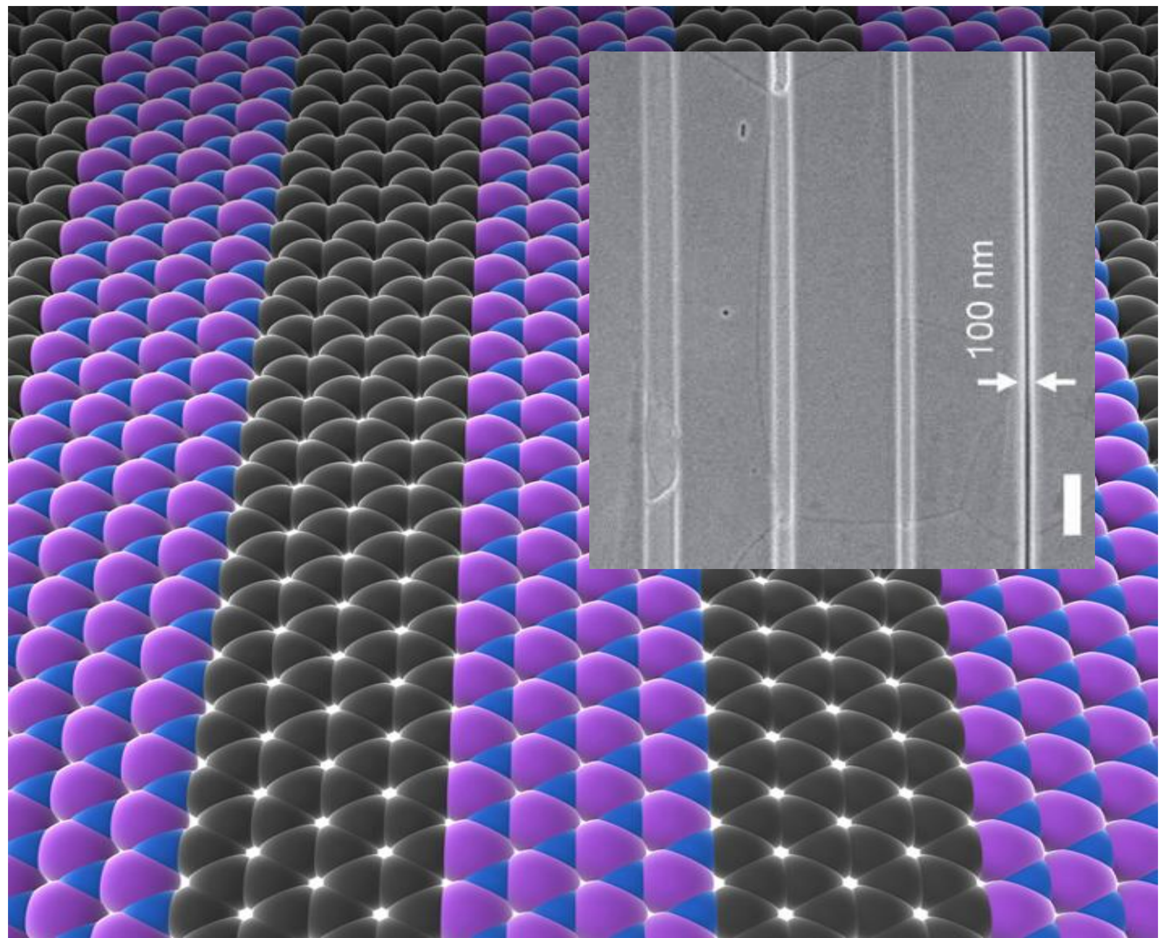


Pulickel Ajayan

Nature Nanotech 8, 119 (2013)

Jiwoong Park

Nature 488, 627–632 (2012)



Multiscale and Predictive modelling

Charge/Thermal/Spin transport

(Kubo, (Spin)-Hall Kubo, Landauer-Büttiker)

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Order-N [Tight-binding-like H]

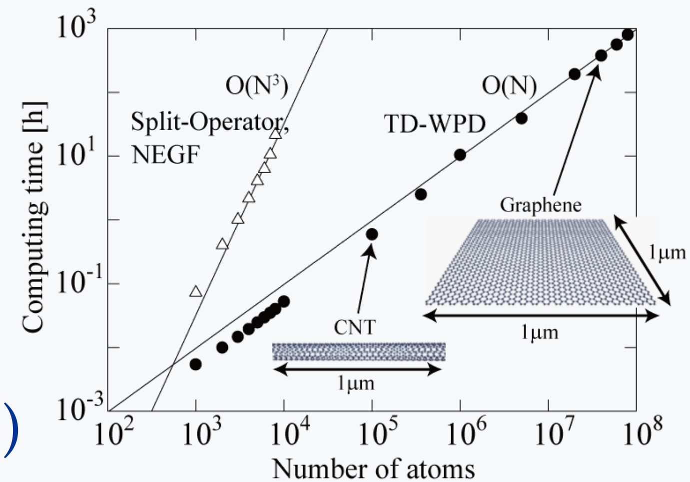
Disorder systems, Magnetic fields,

- Charge transport
- Thermal transport
  (phonon dynamics-harmonic approx.)
- Electron-phonon coupling
  (molecular dynamics, T-dependence)
- Polaron transport (Lang-Firsov Transf.)
- Spin transport (SOC)

$$\sigma_{dc} = e^2 n(E_F) \lim_{t\to\infty} D(t)$$

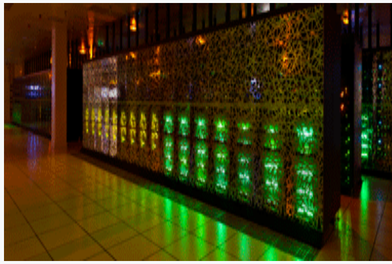
$$D(t) = \frac{\mathrm{Tr}\left[[\hat{X},\hat{U}(t)]^\dagger \delta(E-\mathcal{H})[\hat{X},\hat{U}(t)]\right]}{\mathrm{Tr}[\delta(E-\mathcal{H})]}$$

H. Ishii et al
C.R. Physique 10, 283 (2009)

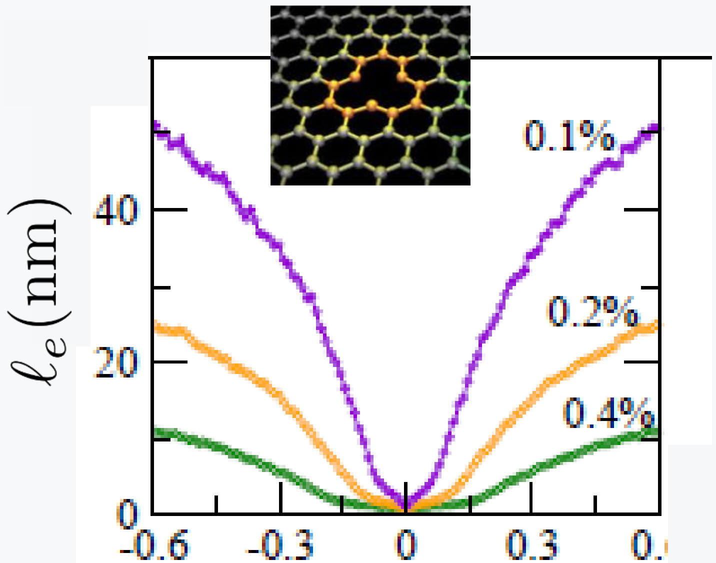


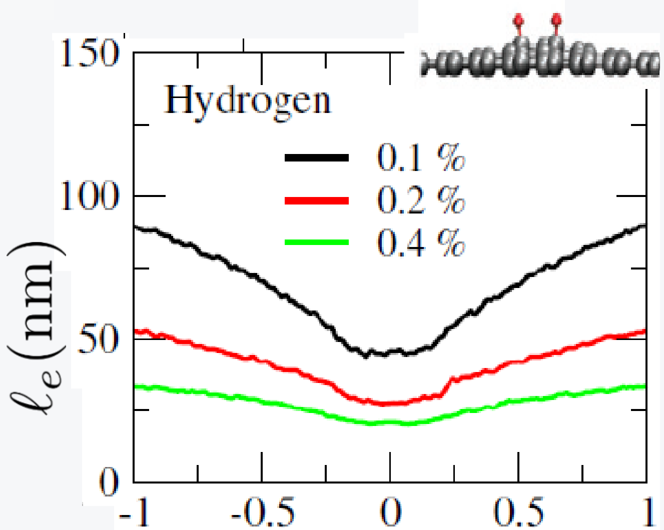
Ab-initio siesta A linear-scaling density-functional method

High-Performance Computing



Mean free path in intentionally damaged graphene

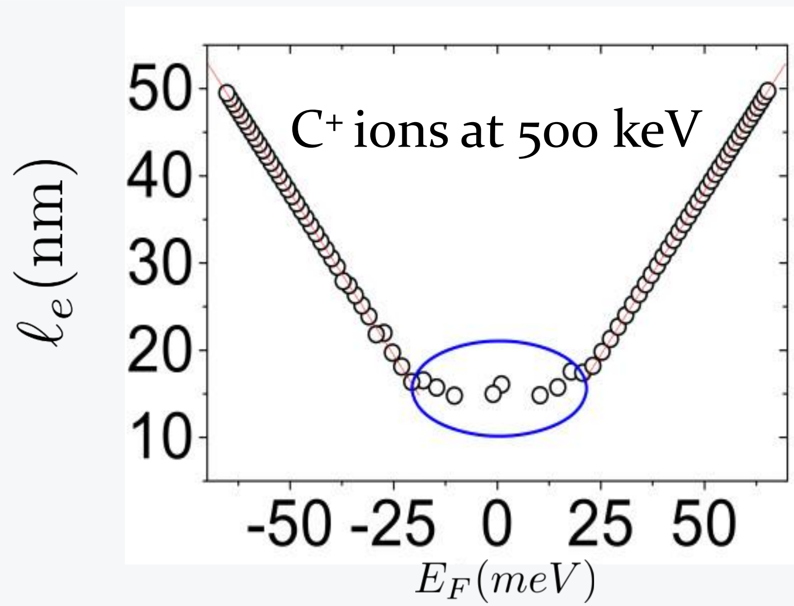




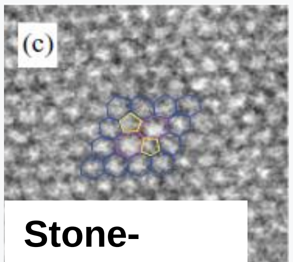
Experimental data :

F. Giannazzo et al,

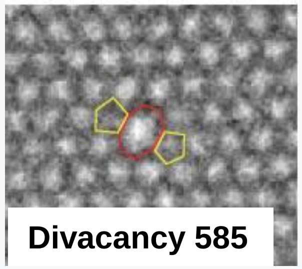
Nanoscale Res.Lett.6, 109 (2011)



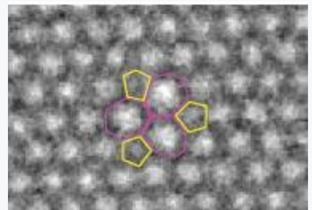
Low mobility & Insulating regime
1% of structural defects



Stone-Wales



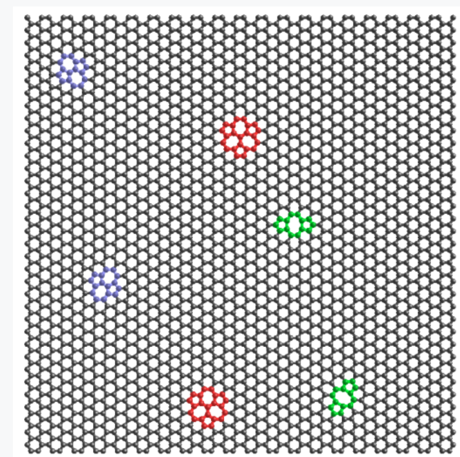
Divacancy 585



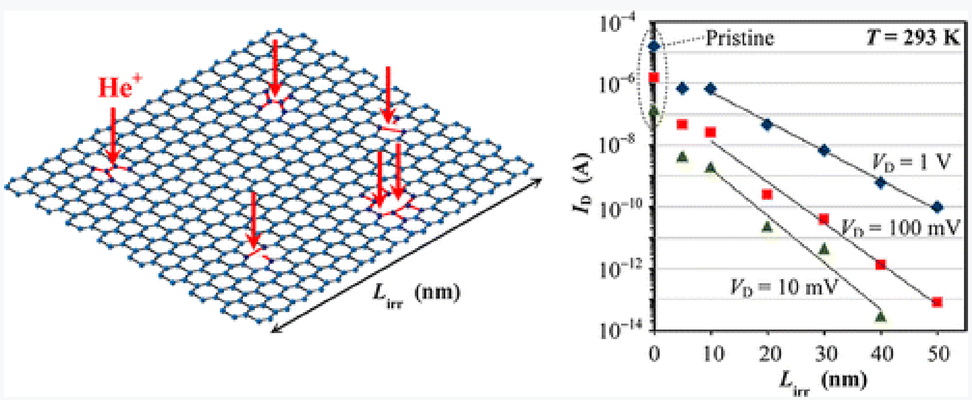
Divacancy 555777

A. Lherbier et al.,
Phys. Rev. Lett 106, 046803 (2011)

For 1% of such defects



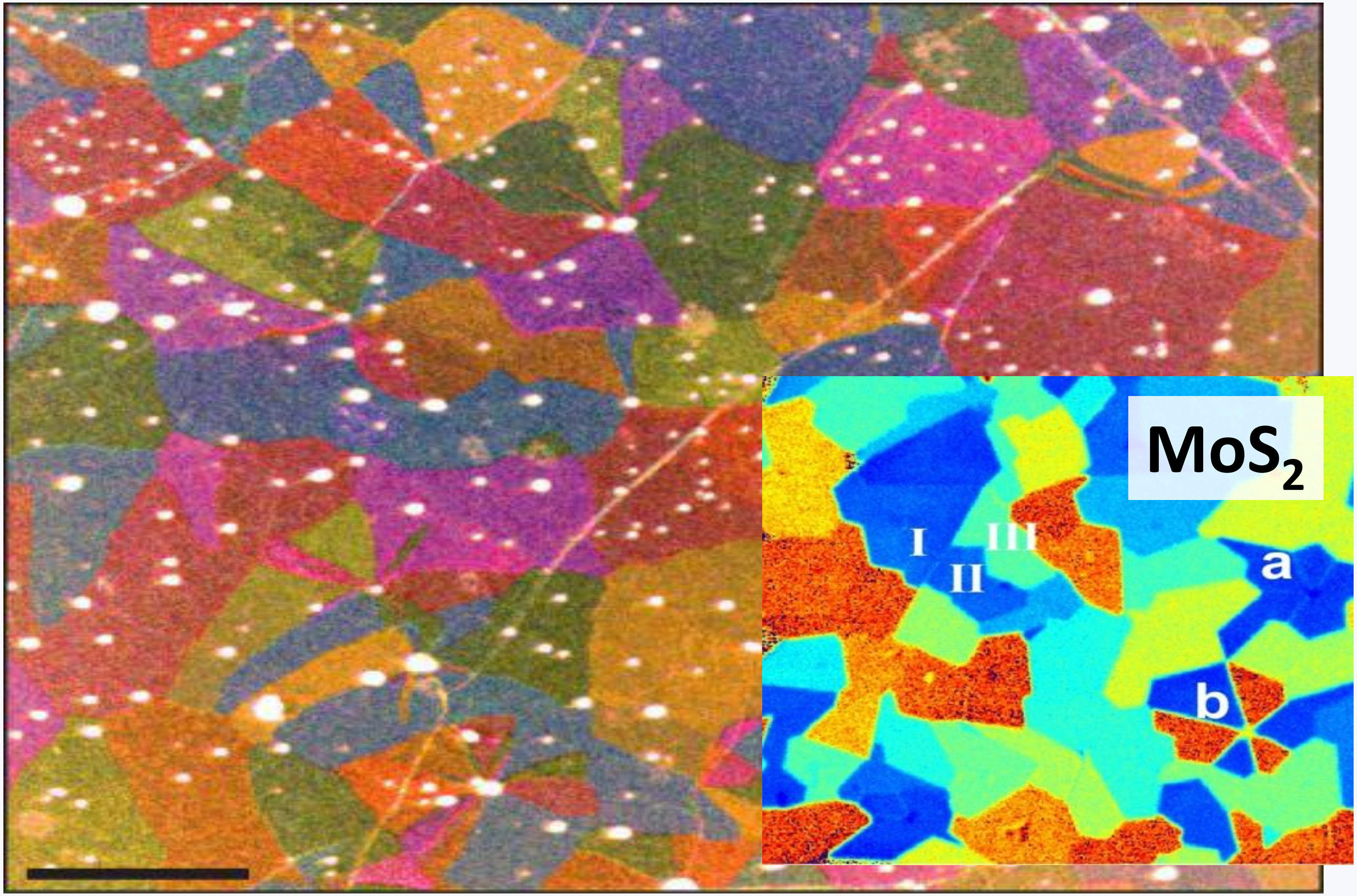
Nakaharai et al.,
ACS Nano 7 (2013) 5694



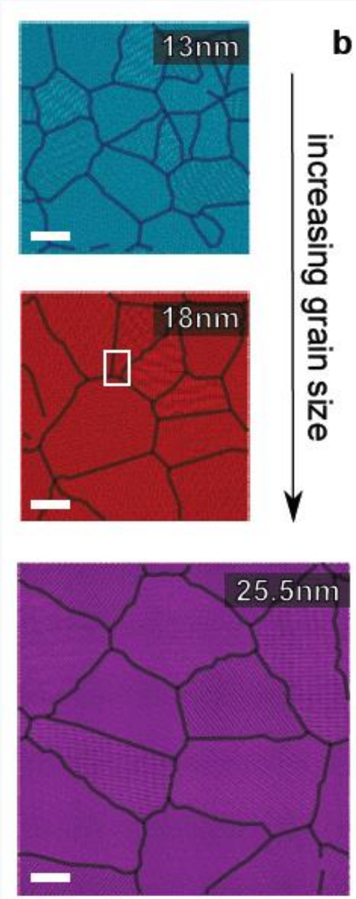
$$\mu \sim 1 - 10 \mathrm{cm}^2 \mathrm{V}^{-1} \mathrm{s}^{-1}$$

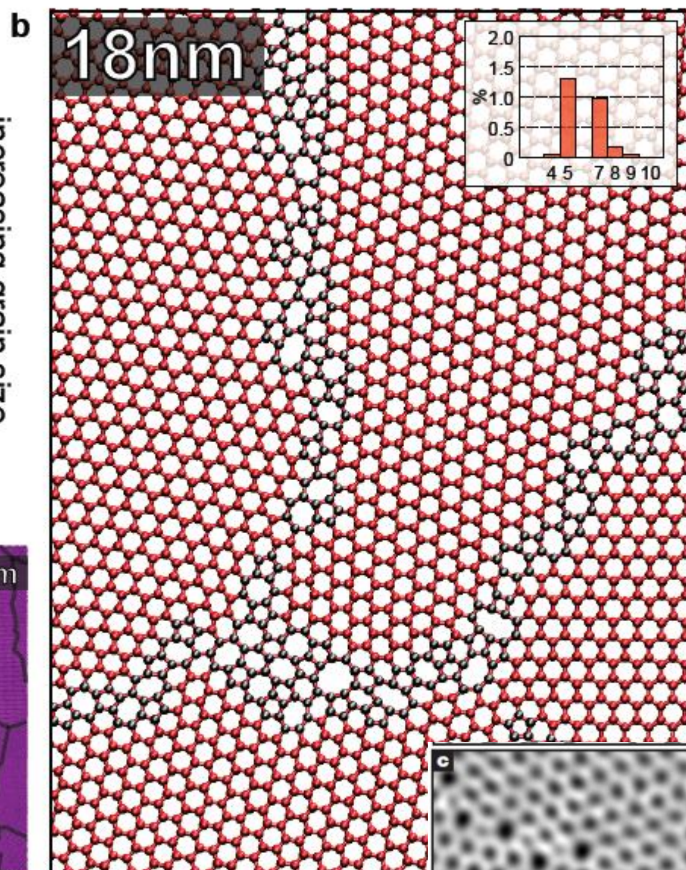
$$\xi \sim 10 \mathrm{nm}$$

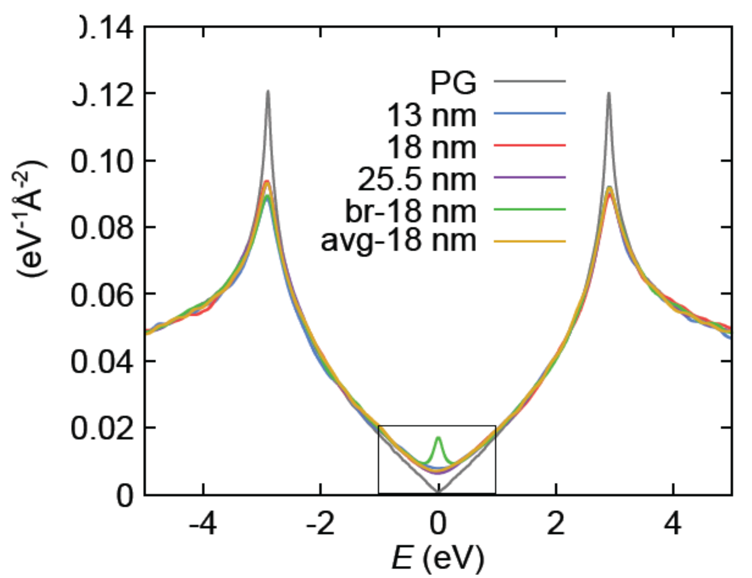
Polycrystalline Graphene

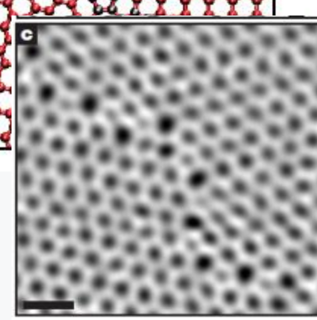


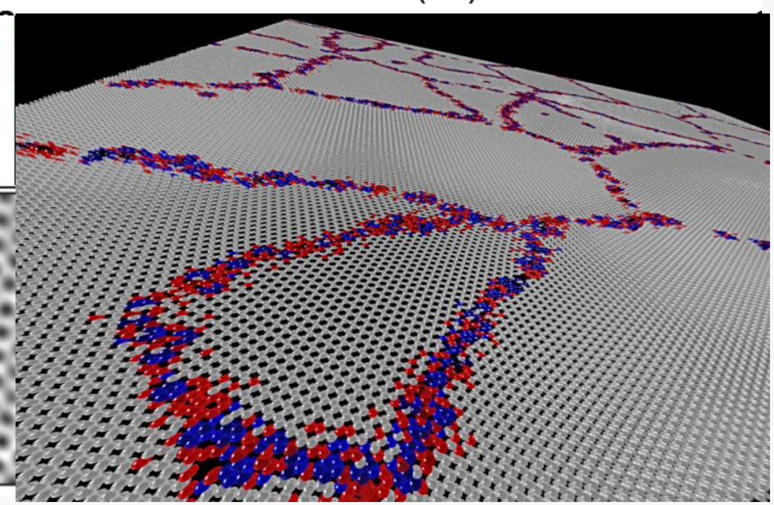
Polycrystalline graphene (models)





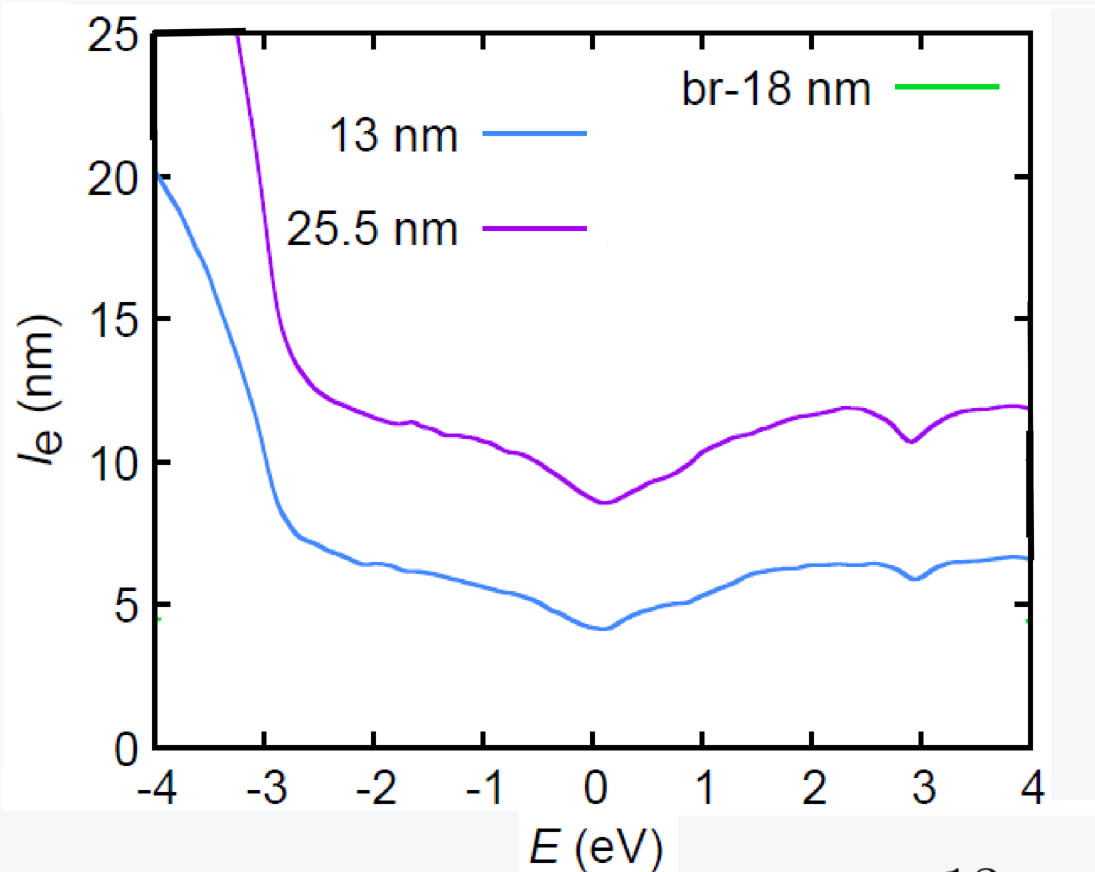


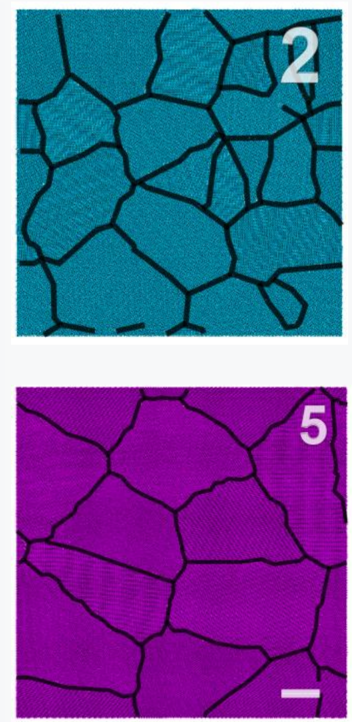




Coll.
Jani Kotakoski

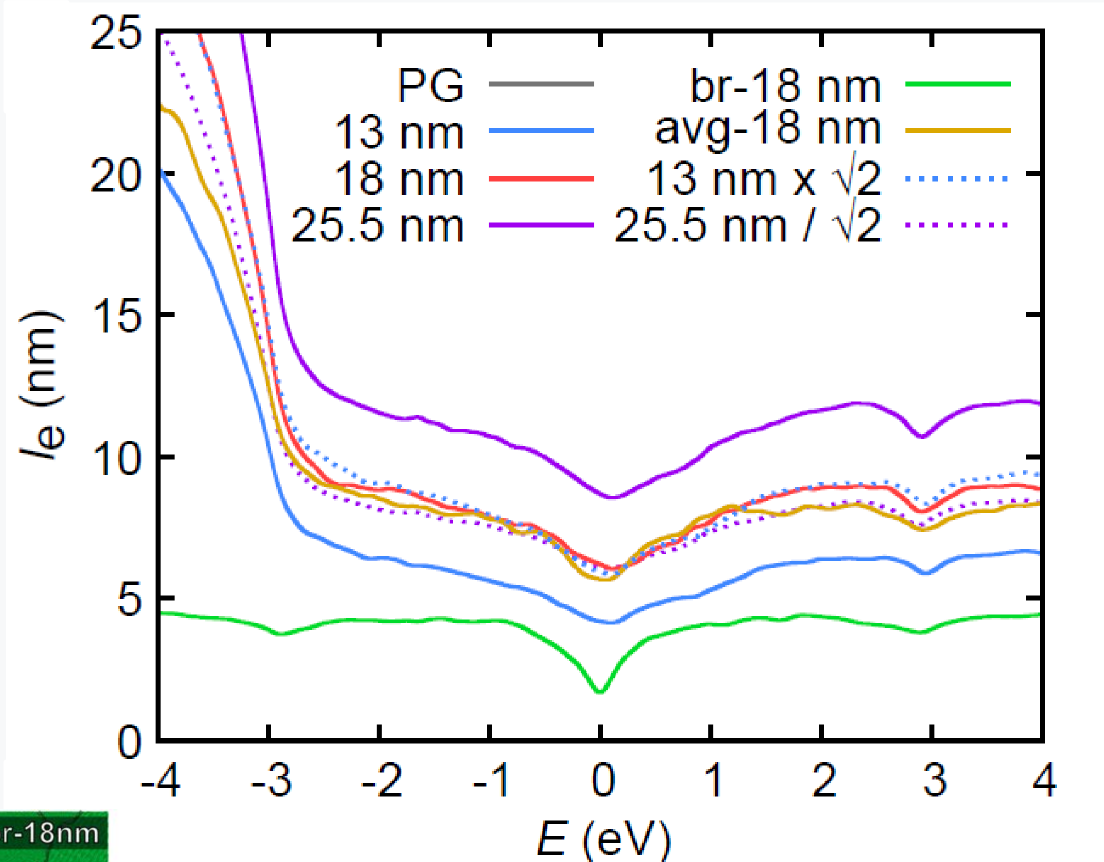
Elastic mean free path

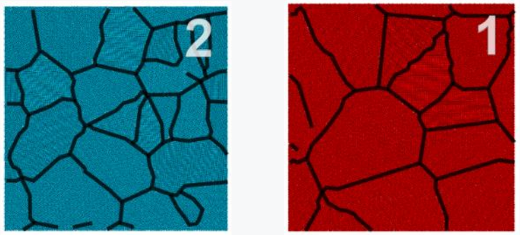




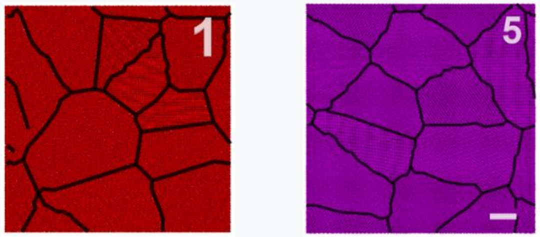
$$\ell_e^{13\text{nm}} \times 2 \simeq \ell_e^{25.5\text{nm}}$$

Transport scaling law (well connected grains)



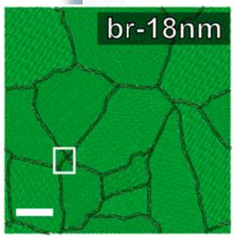


$$\sqrt{2} \times \ell_e^{13\mathrm{nm}} \approx \ell_e^{18\mathrm{nm}}$$



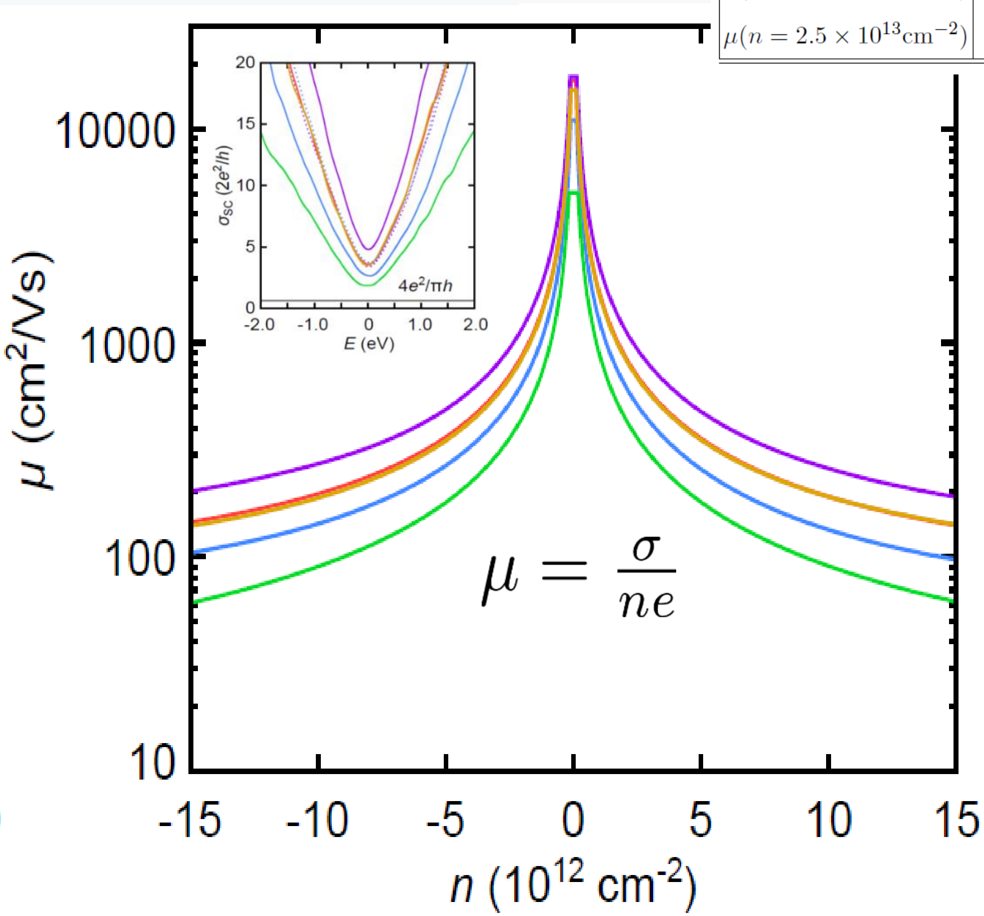
$$\sqrt{2} \times \ell_e^{18\ \mathrm{nm}} \approx \ell_e^{25.5\mathrm{nm}}$$

$$\ell_e \sim d_{\mathrm{grainsize}}$$



Charge Mobility of polyG

Mobilities (cm^2/Vs)	13 nm	18nm	avg-18nm	25.5nm	br-18nm
$\mu(n = 2.5 \times 10^{11} cm^{-2})$	5.1×10^3	7×10^3	6.8×10^3	10^4	4×10^3
$\mu(n = 2.5 \times 10^{12} cm^{-2})$	510	700	685	950	360
$\mu(n = 2.5 \times 10^{13} cm^{-2})$	69	105	104	150	45



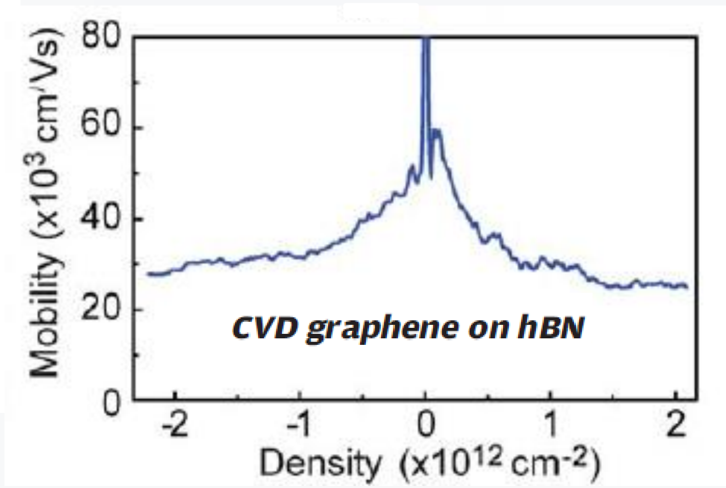
$$\mu = \frac{\sigma}{ne}$$

Extrapolation

$$d_{grainsize} \sim 1\mu m$$

$$n = 3 \times 10^{11} cm^{-2}$$

$$\mu \sim 300.000 cm^2 V^{-1} s^{-1}$$

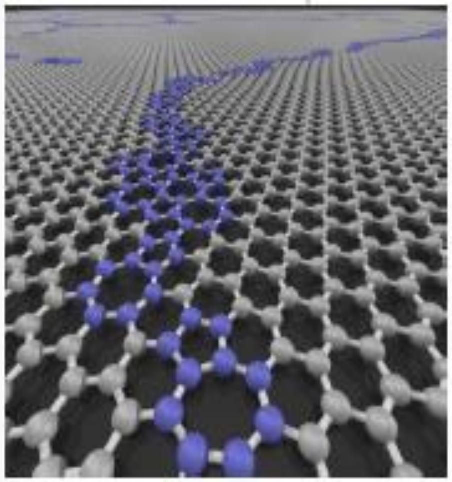


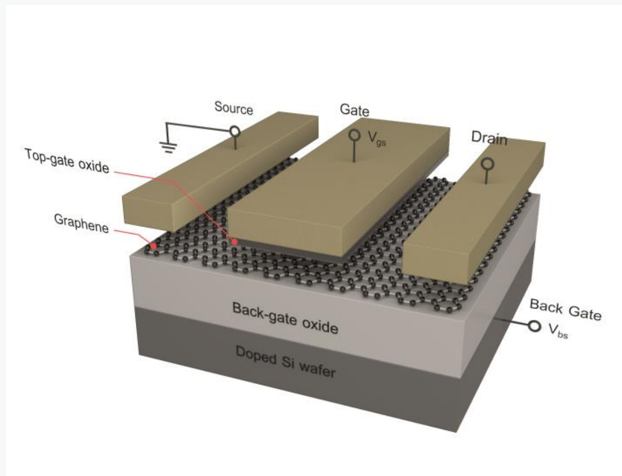
D.V.Tuan et al Nano Letters 13, 1730 (2013)
Petrone et al., Nano Letters , 12 ,2751 – 2756 (2012)

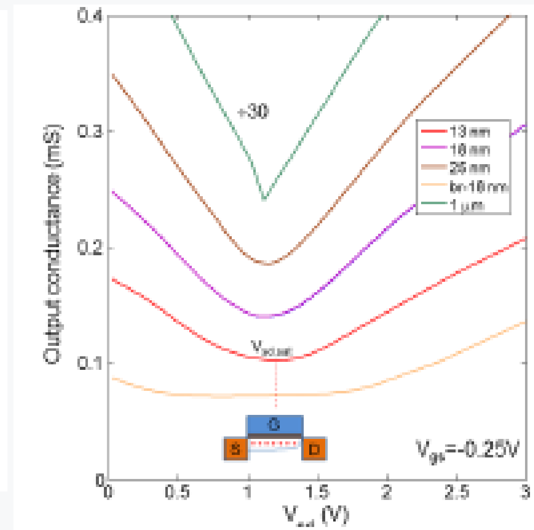
Multiscale Graphene device simulation

Institut Català de Nanociència i Nanotecnologia

Modeling of Graphene-FETs Drift-diffusion Transport







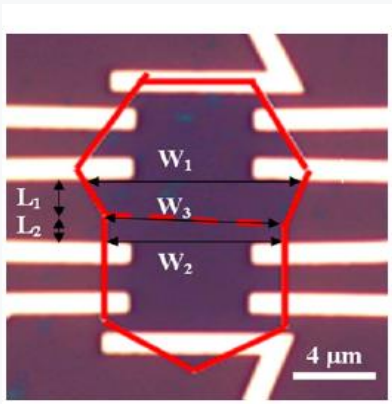
Model parameters extracted from intrinsic material properties (charge mobility)

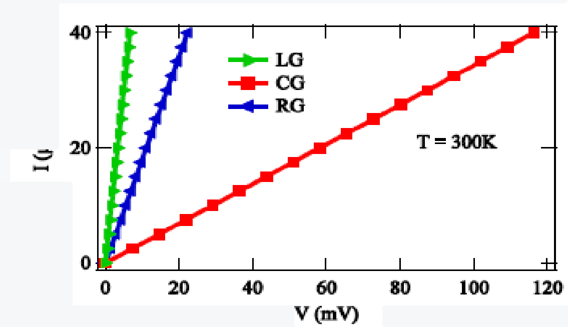
$$I_{ds} = \frac{\mu W \int_0^{V_{ds}} |Q_c| dV}{L + \mu \frac{|V_{ds}|}{v_F}}$$

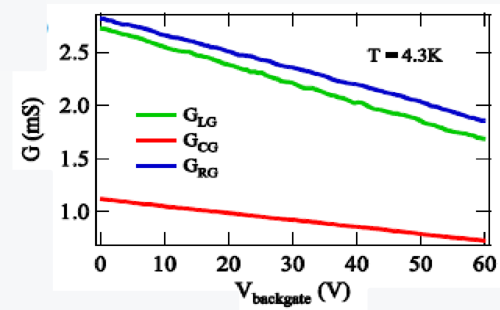
SAMSUNG

D. Jiménez, A. Cummings, F. Chaves,
D. Van Tuan, J. Kotakoski, S. Roche,
Applied Physics Letter 104, 043509 (2014)

Comparison with experiments



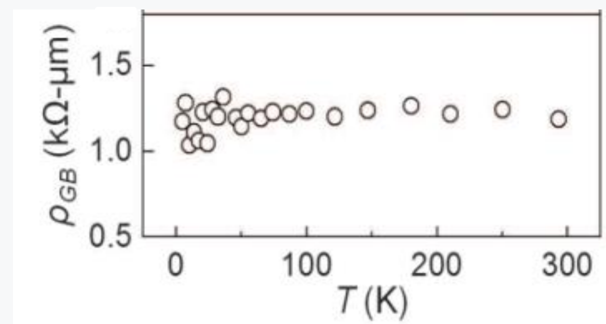




Intra-grain conductance and **inter-grain resistance** R_{GB}

Effective intergrain resistivity ρ_{GB}

$$\rho_{GB}^{expt.} \in [0.5, 8]\mathrm{k}\Omega.\mu\mathrm{m}$$

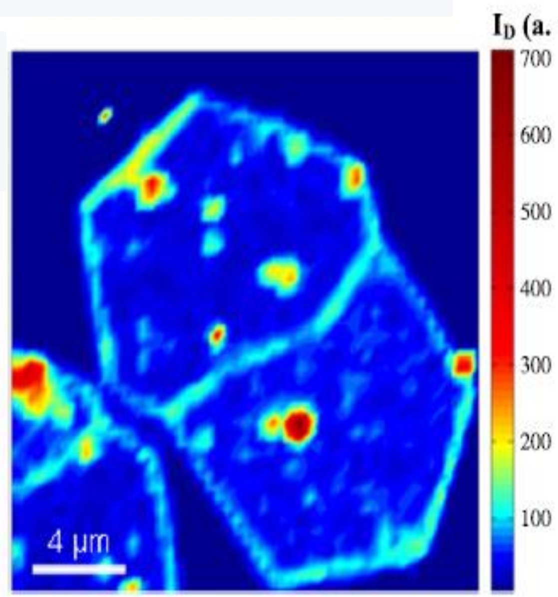


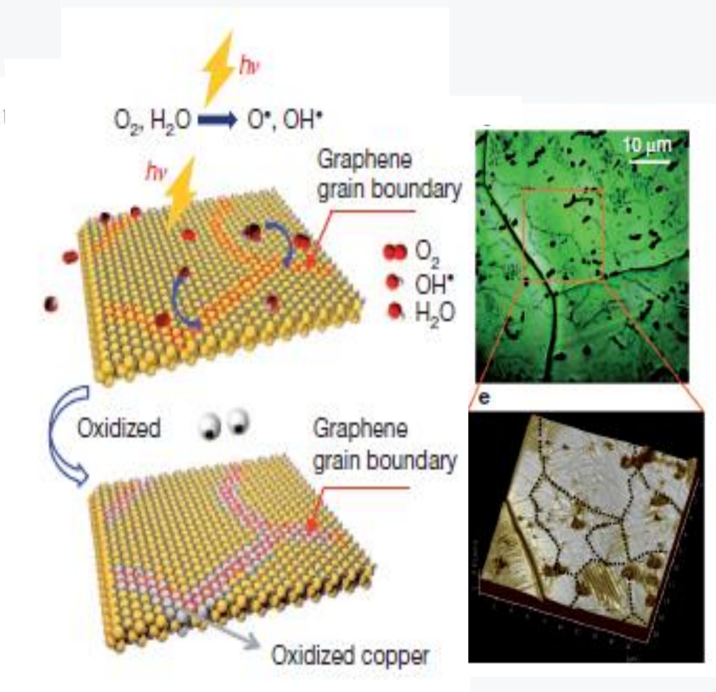
Jauregui et al, Solid. Stat Comm. 151, 1100(2011)
Yu et al, Nat. Mater. 10, 443 (2011)
Tsen et al, Science 336, 1143 (2012)
Duong et al. Nature 490, 235 (2012)

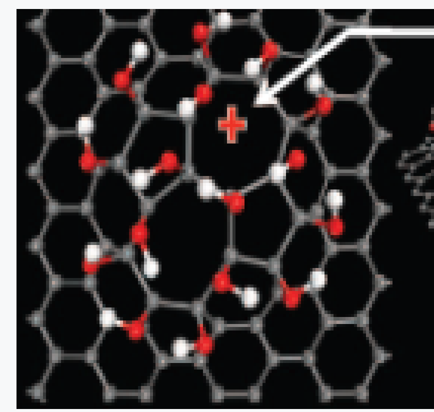
$$\rho_{GB}^{simul.} \sim 0.065\mathrm{k}\Omega.\mu\mathrm{m}$$

Origin of lower values in experiments?

Duong et al. Nature 490, 235 (2012)
Young-Hee Lee (SKKU)

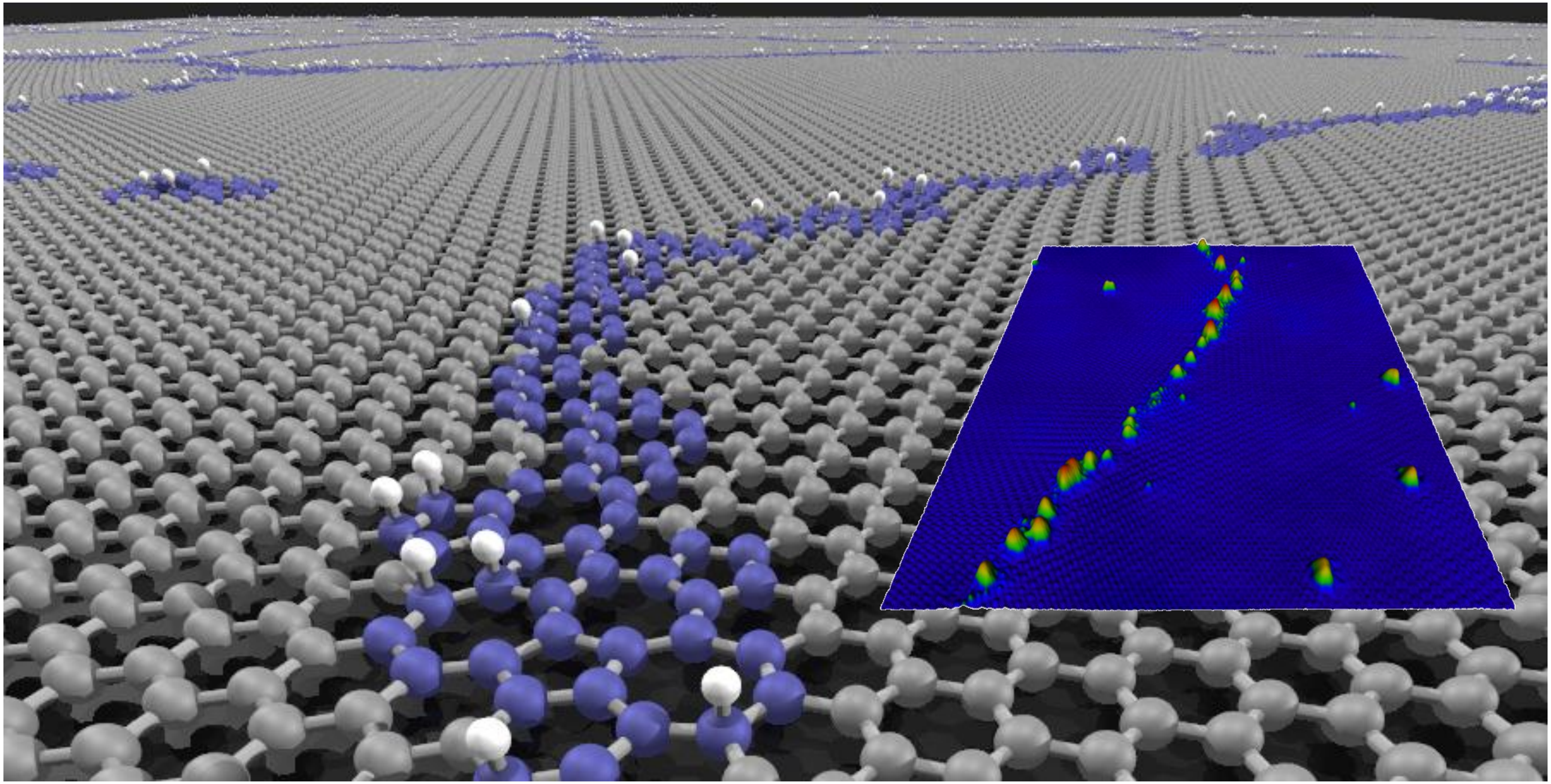






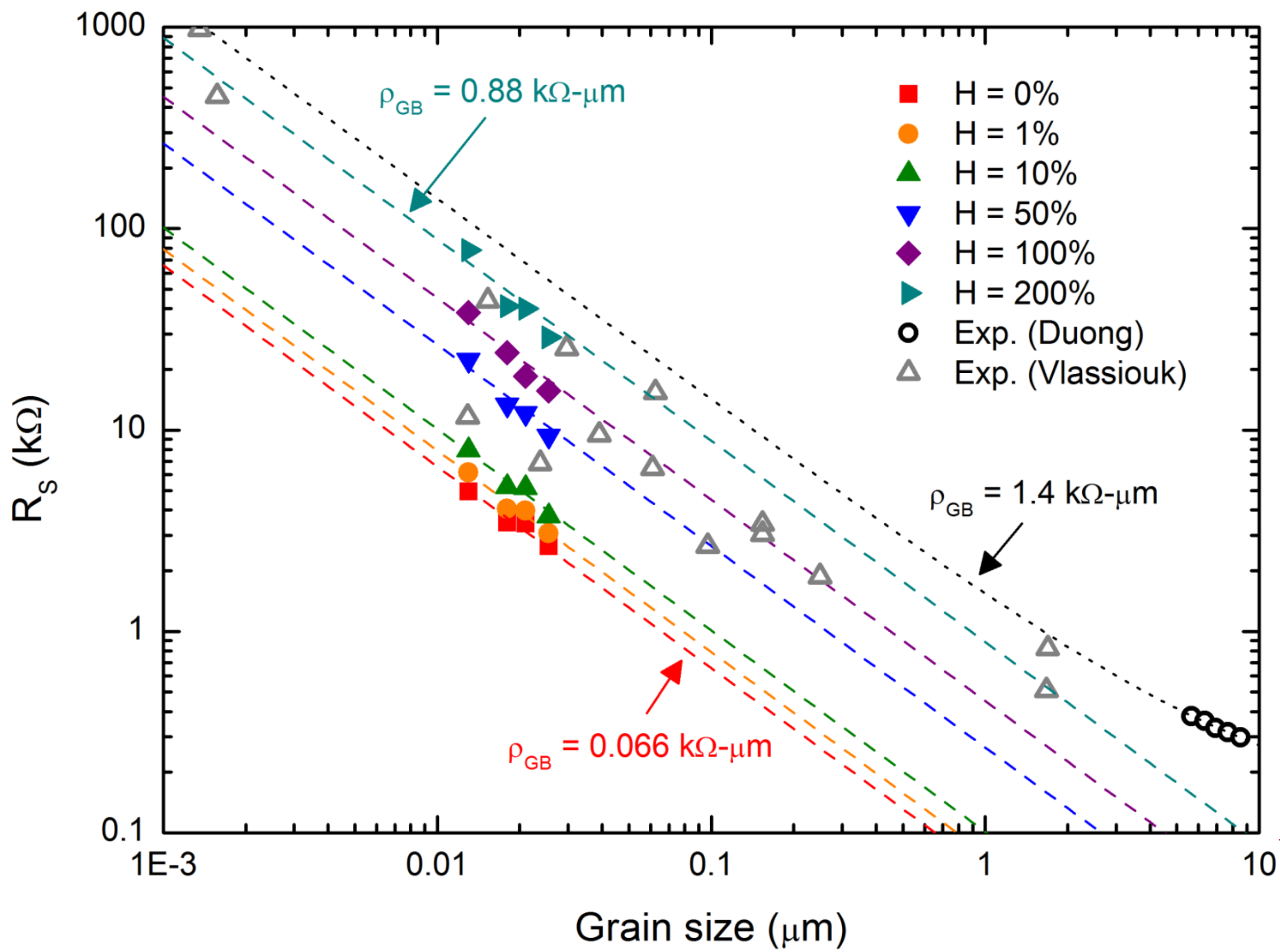
Chemical functionalization of grain boundaries

*Adsorbates randomly placed (**only**)on GB atoms*



Resistance scaling upon adsorption

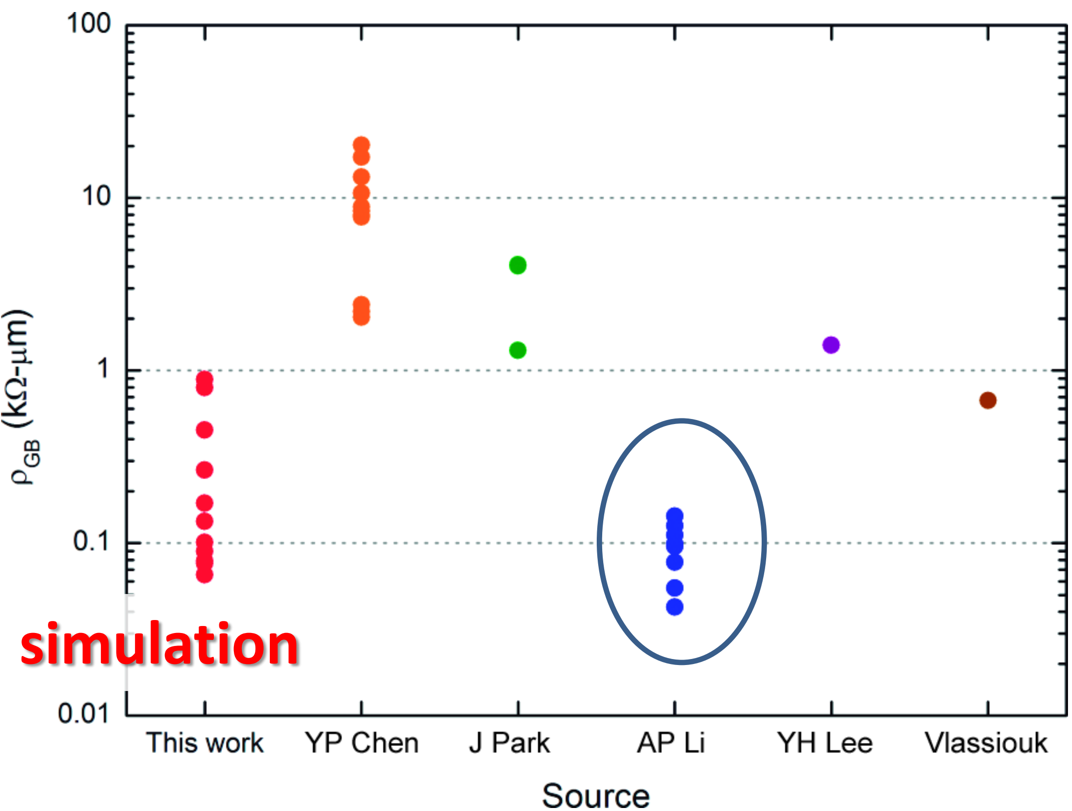
Sheet resistance vs. grain size



Technological target

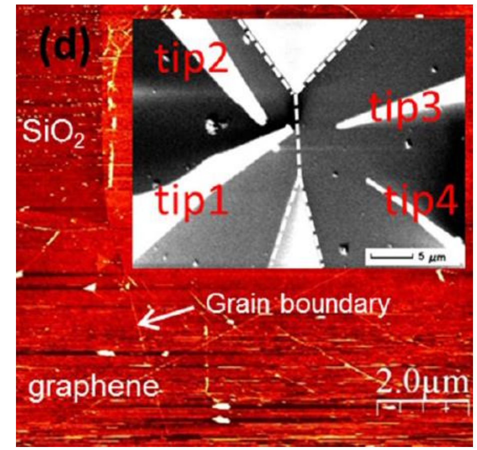
Significant increase in R_S with GB adsorbates

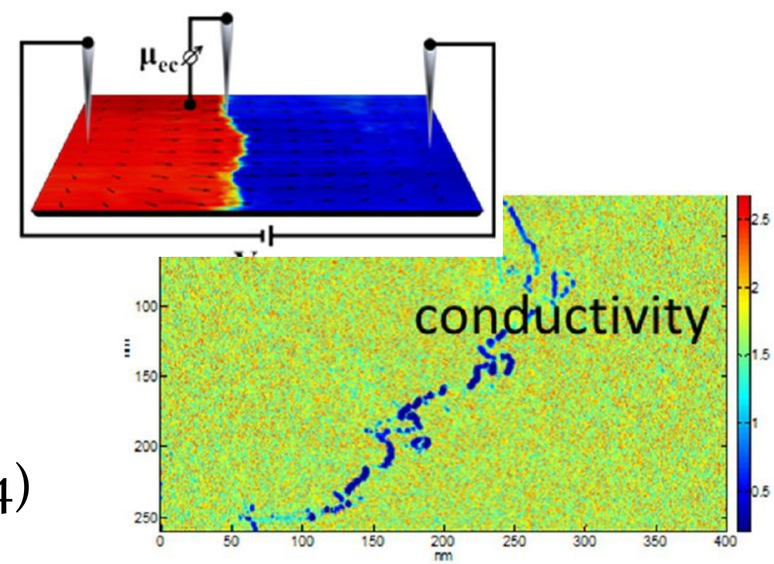
Comparison with reported experimental data



A. Cummings et al,
Advanced Materials 26, 5079–5094 (2014)

4-probe STM probe
Clark etal
ACS Nano, 2013, 7 (9), pp 7956–796





GB resistivity vs. adsorbate nature and concentration

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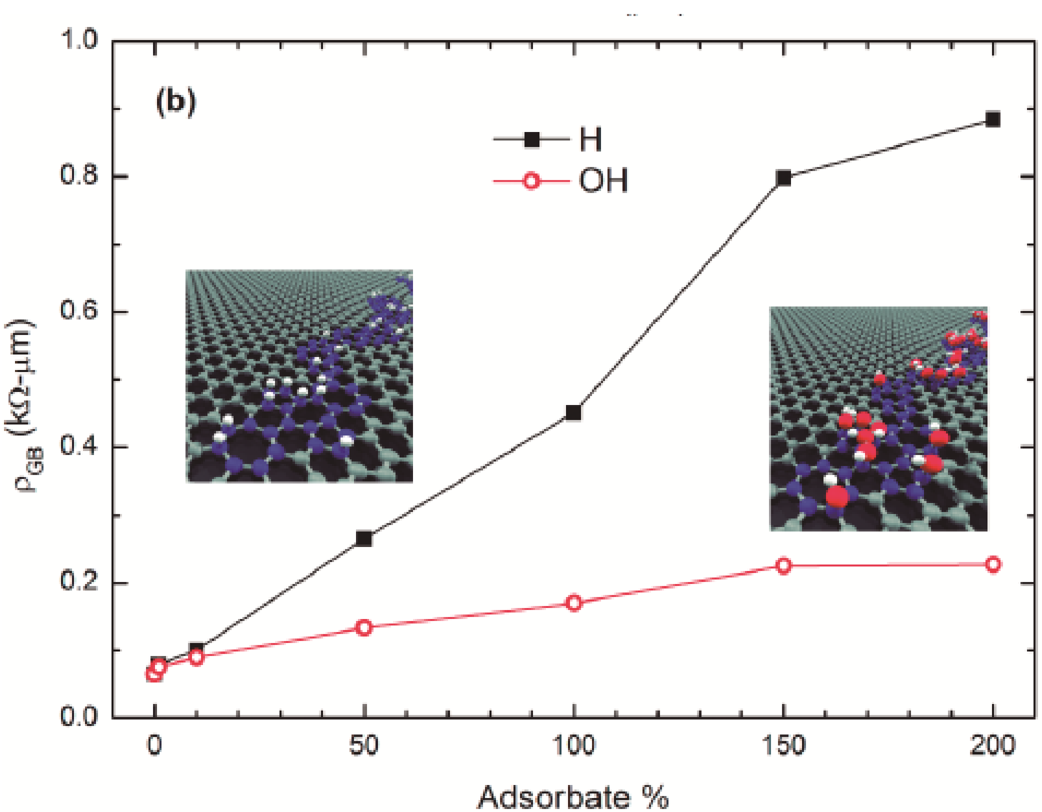
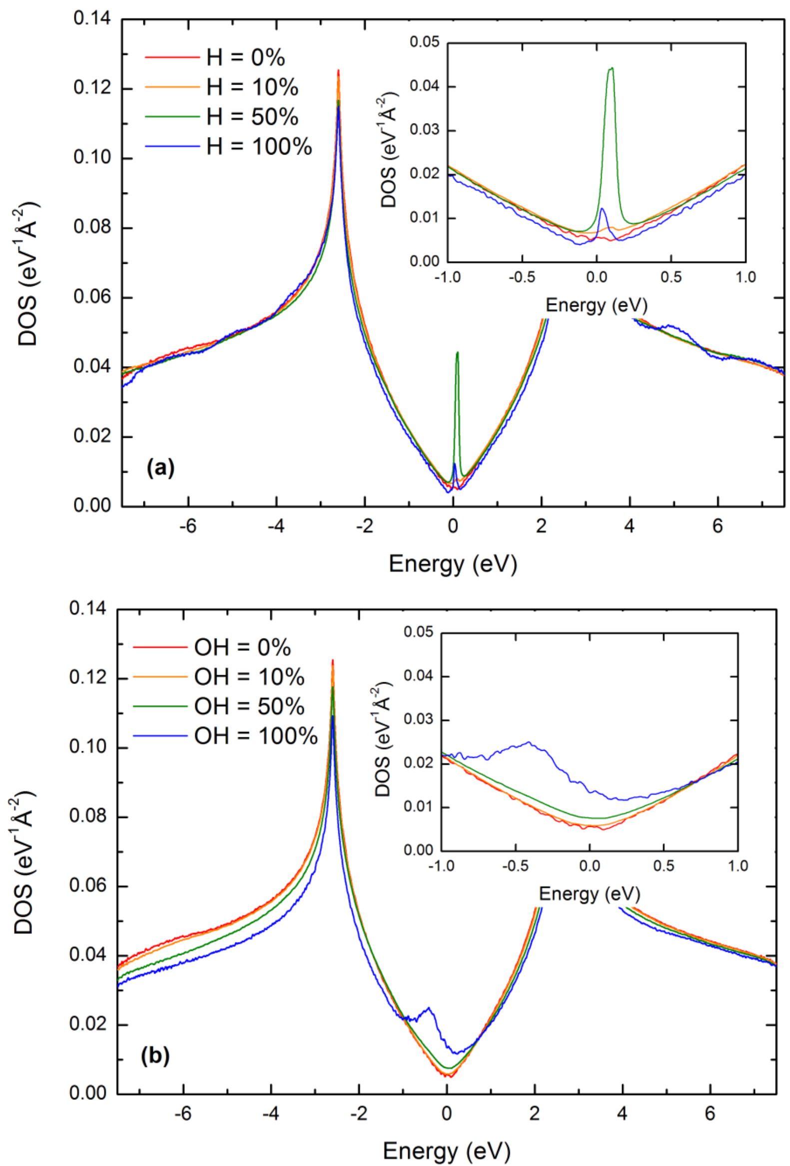


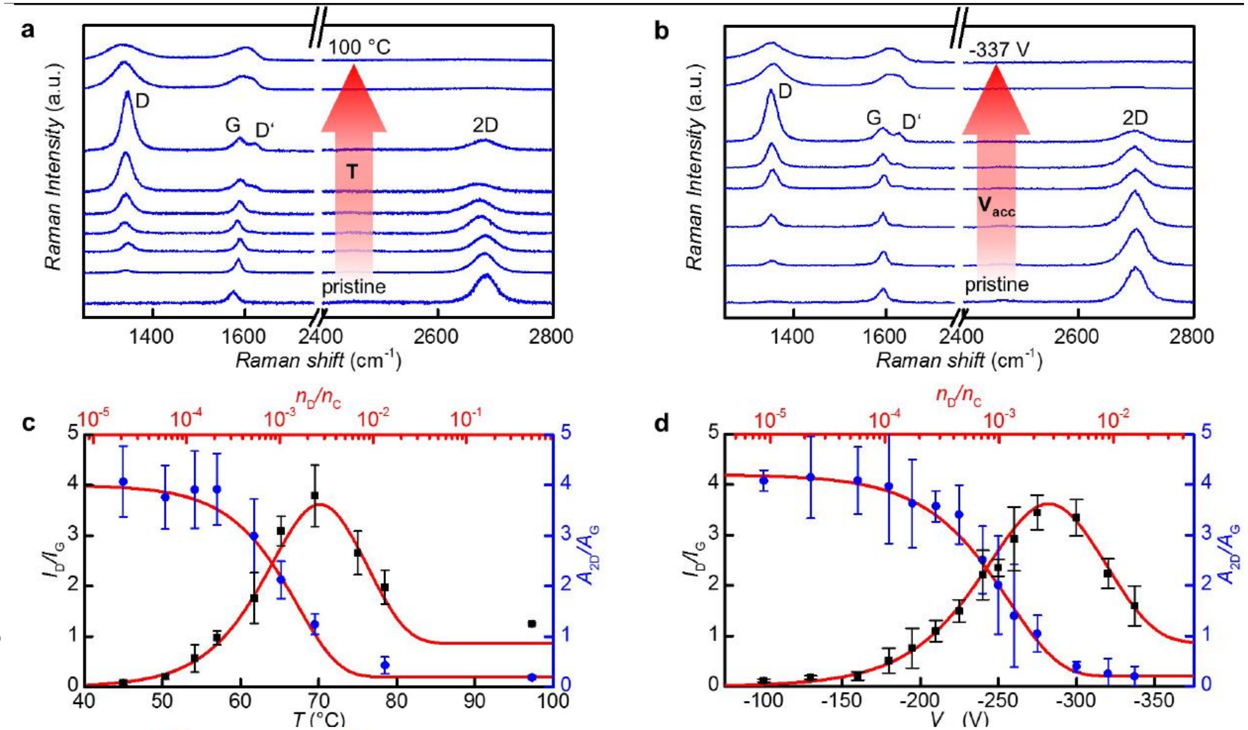


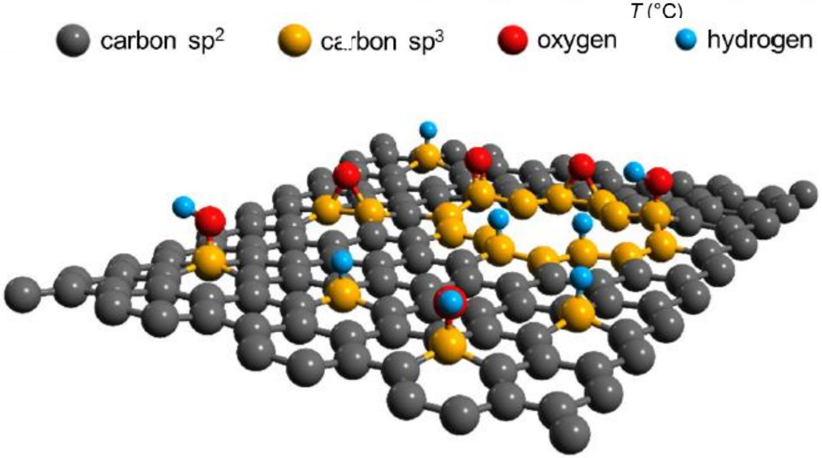
Localized impurity band near Dirac point (H)

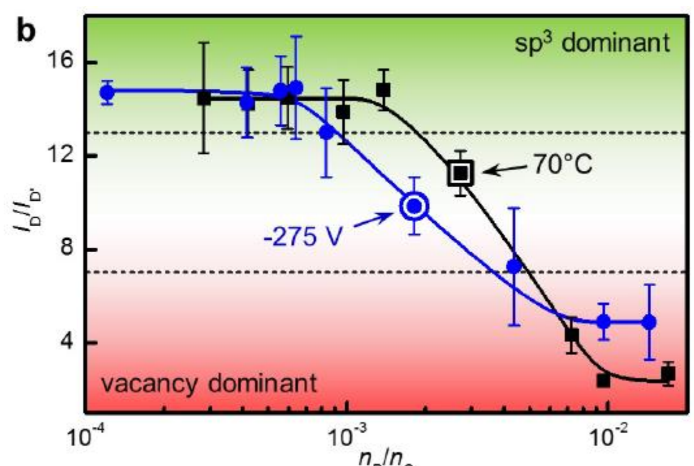
Dispersive impurity band (OH)

Controlled oxidation and hydrogenation of CVD graphene

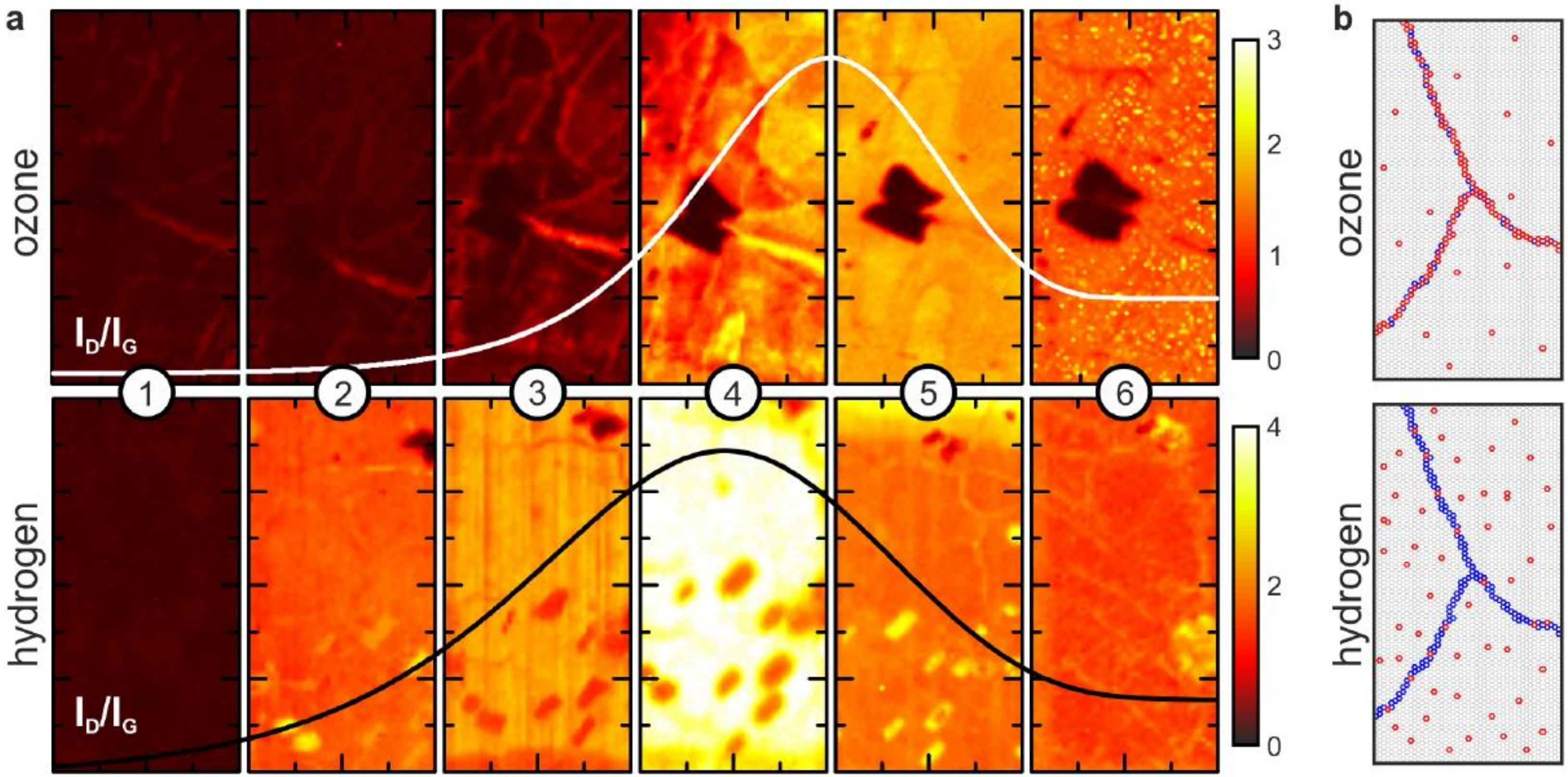
José-Antonio Garrido







Controlled oxidation and hydrogenation of CVD graphene



Conclusions : OZONIZATION

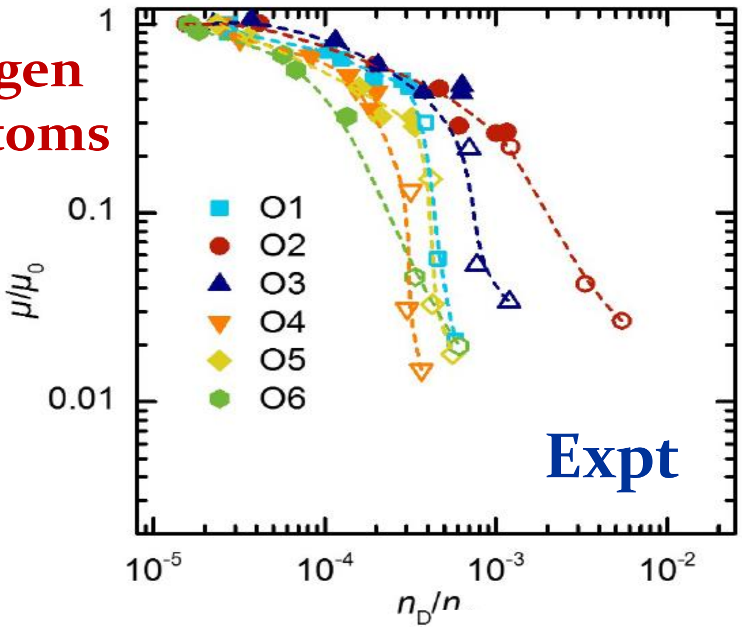
Formation of epoxide, O-related defects first massively populate Grain boundaries. After saturation limit, cracks ("vacancy") are created around GBs (etching graphene)

HYDROGENATION

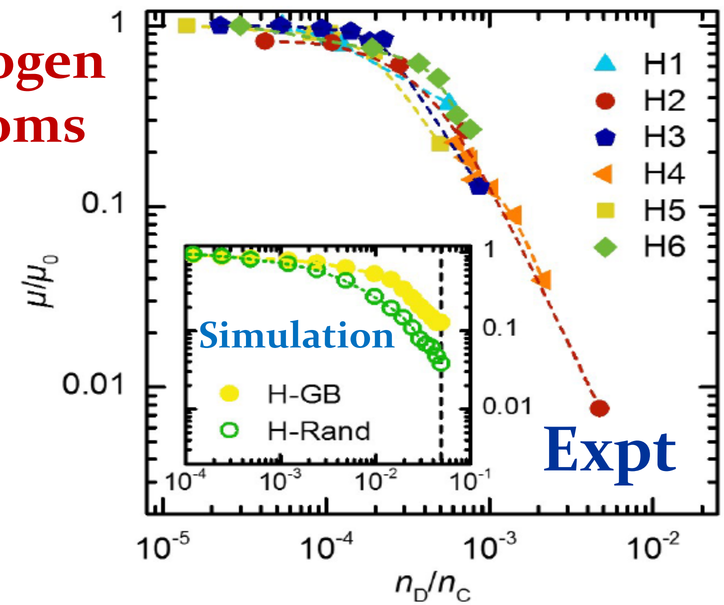
Hydrogen ad-atoms are homogeneously distributed. At a certain density presence of vacancies start dominate the Raman features

Charge mobility scaling

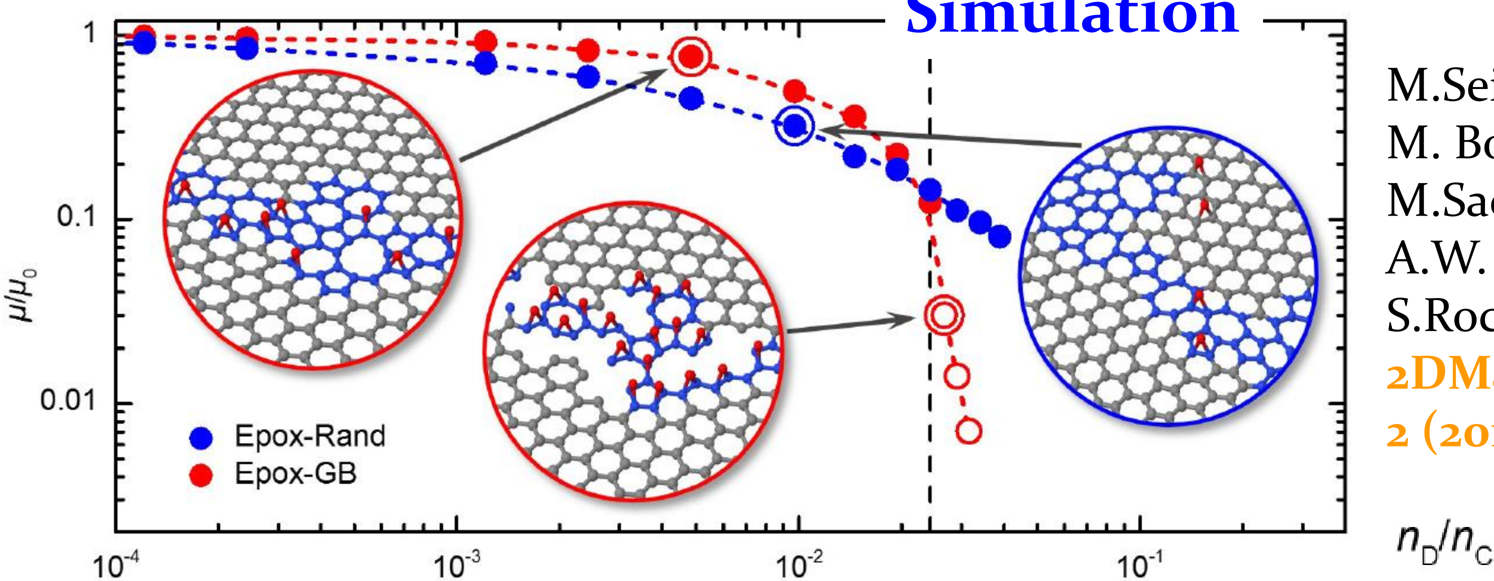
Oxygen adatoms



Hydrogen adatoms



Simulation



M.Seifert, J. Vargas, M. Bobinger, M.Sachsenhauser, A.W. Cummings, S.Roche and J. A. Garrido
2DMaterials 2 (2015) 024008

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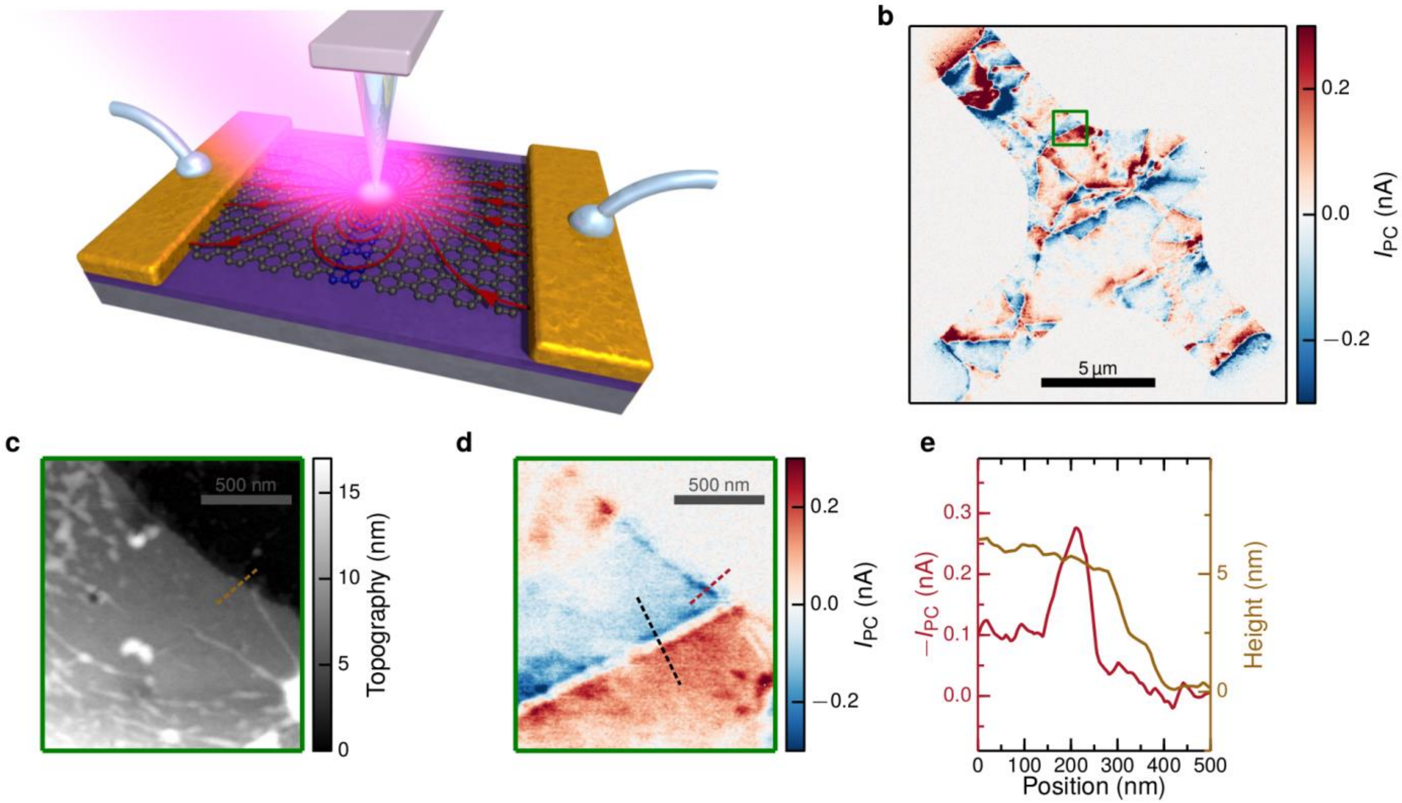
Space dependent photocurrent measurements to characterize polycrystalline graphene

ICFO
Institut de Ciències Fotòniques

Frank Koppens



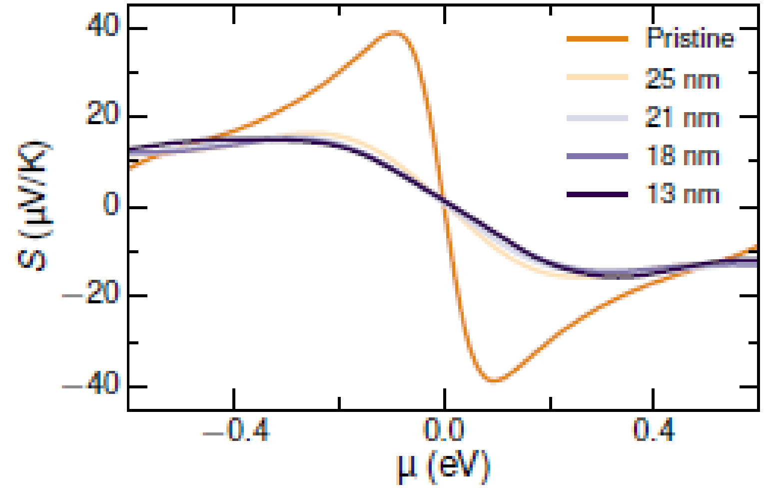
Near-field photocurrent nanoscopy on bare and encapsulated graphene

A.Woessner, P.Alonso-González, M. B. Lundeberg, Y. Gao, J. E. Barrios-Vargas, G.Navickaite, Q.Ma, D. Janner, K.Watanabe, A. W. Cummings, T. Taniguchi, V.Pruneri, S. Roche, P. Jarillo-Herrero, J.Hone, R. Hillenbrand, F.H.L. Koppens

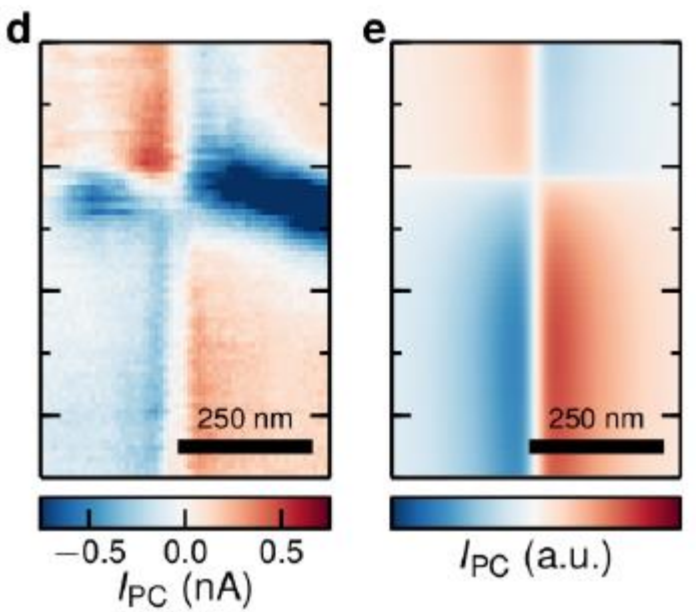
arXiv:1508.07864

Simulation of Seebeck coefficients in Polycrystalline graphene

$$S(\mu,T) = -\frac{1}{|e|T}\frac{\int_{-\infty}^{\infty}(E-\mu)G(E)\left(-\frac{\partial f}{\partial E}\right)dE}{\int_{-\infty}^{\infty}G(E)\left(-\frac{\partial f}{\partial E}\right)dE},$$



$$I_{\mathrm{PC}}(x,y) = \frac{1}{RW}\int \frac{\partial T(x,y)}{\partial x} S(x,y)\, dx dy$$



A.Woessner, F.H.L. Koppens et al

arXiv:1508.07864

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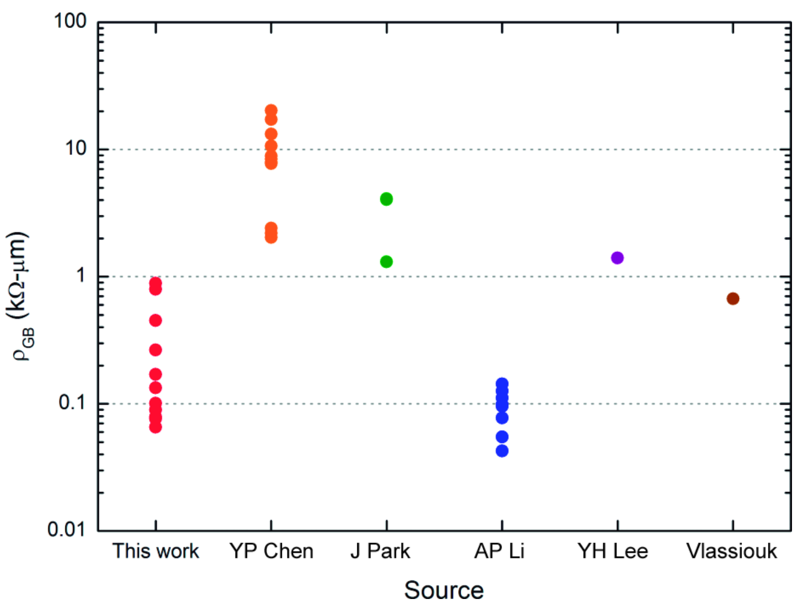
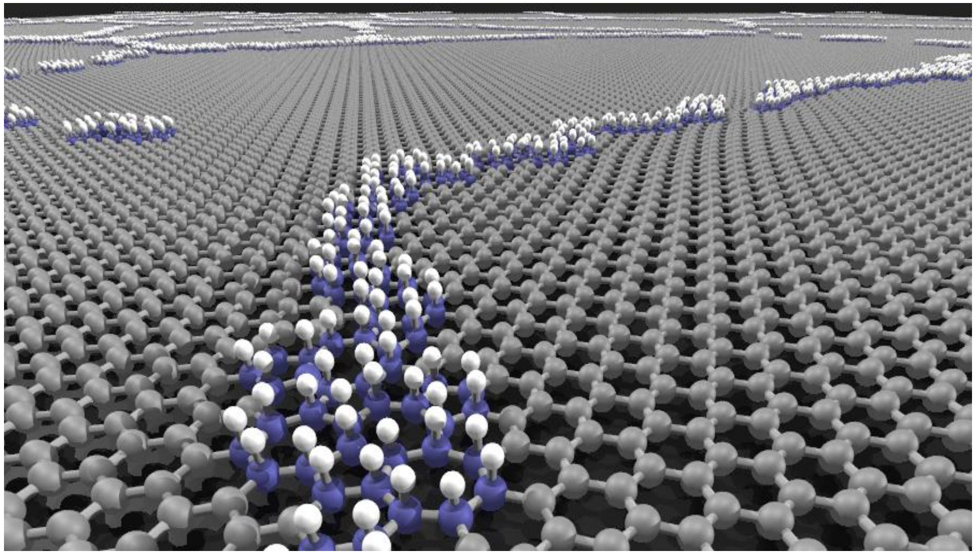
Charge Transport in Polycrystalline Graphene: Challenges and Opportunities

Aron W. Cummings, Dinh Loc Duong, Van Luan Nguyen, Dinh Van Tuan, Jani Kotakoski, Jose Eduardo Barrios Vargas, Young Hee Lee, and Stephan Roche**





Advanced Materials 26, 5079–5094 (2014)

Aknowledgments

GRAPHENE FLAGSHIP

Institut Català de Nanociència i Nanotecnologia

T. Dinh **Eduardo Barrios** **David Soriano** **Aron Cummings**

Collaboration (Experiment)

Young Hee Lee (SKKU)
José-Antonio Garrido (ICN2)
Frank Koppens (ICFO)