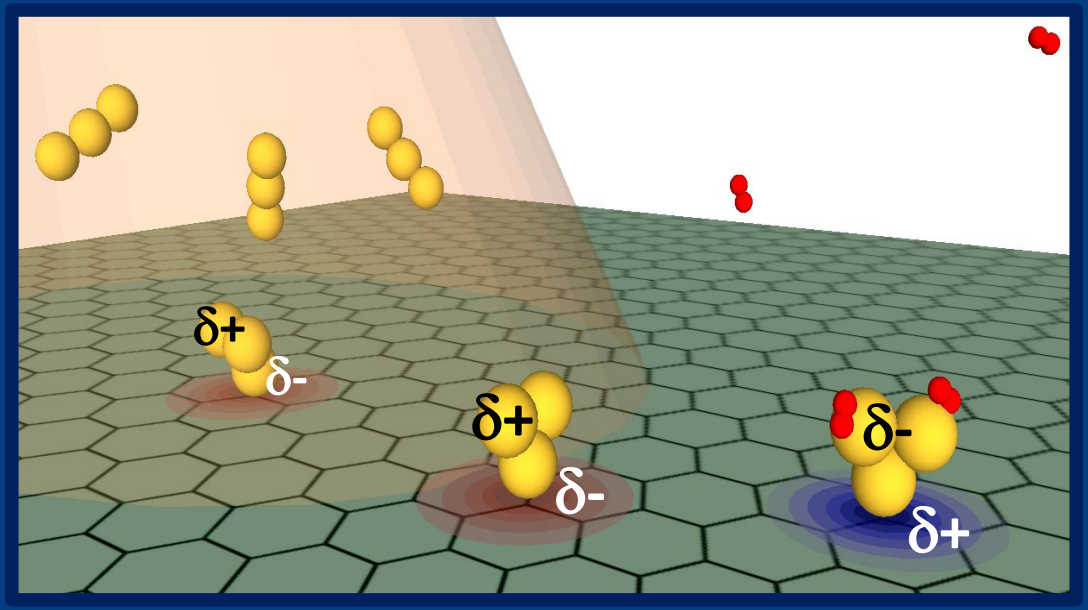
Graphene devices decorated with few-atom clusters

probing and exploiting the size-specific interaction



Jeroen E. Scheerder

Graphene 2018
June 26 - 29
Dresden (Germany)

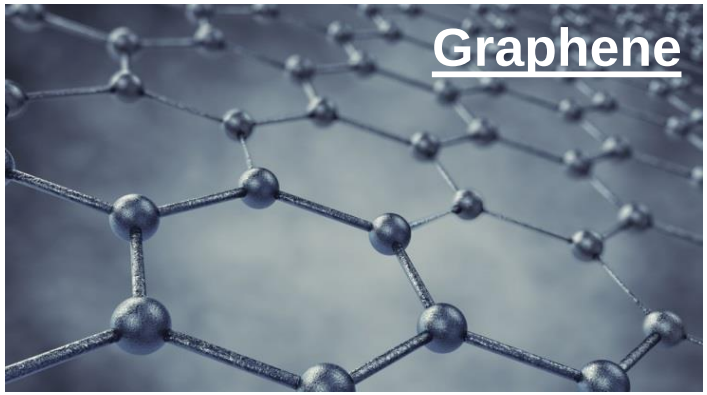
Supervisor: **prof. dr. Joris Van de Vondel**
Co-supervisor: **prof. dr. Ewald Janssens**

KU LEUVEN

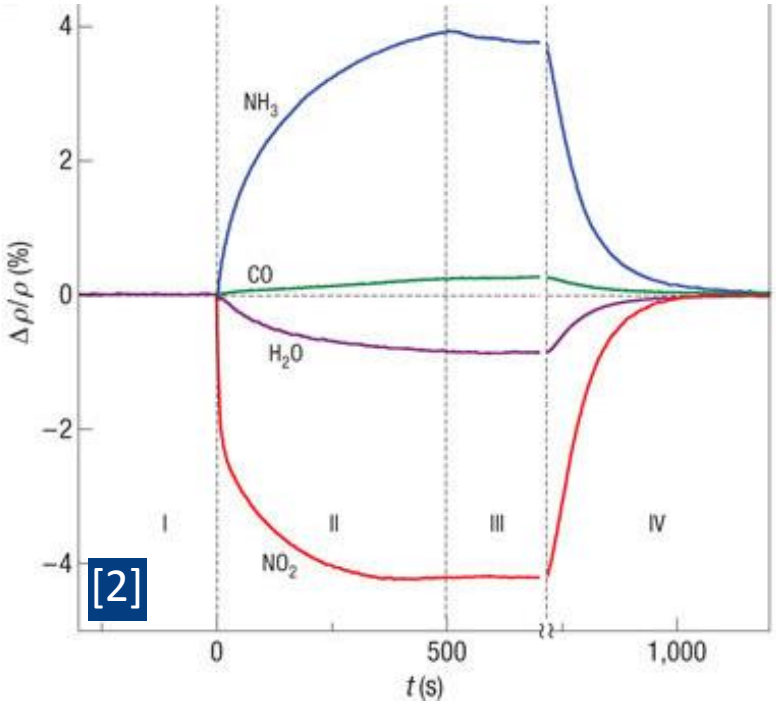
28th June 2018

The framework

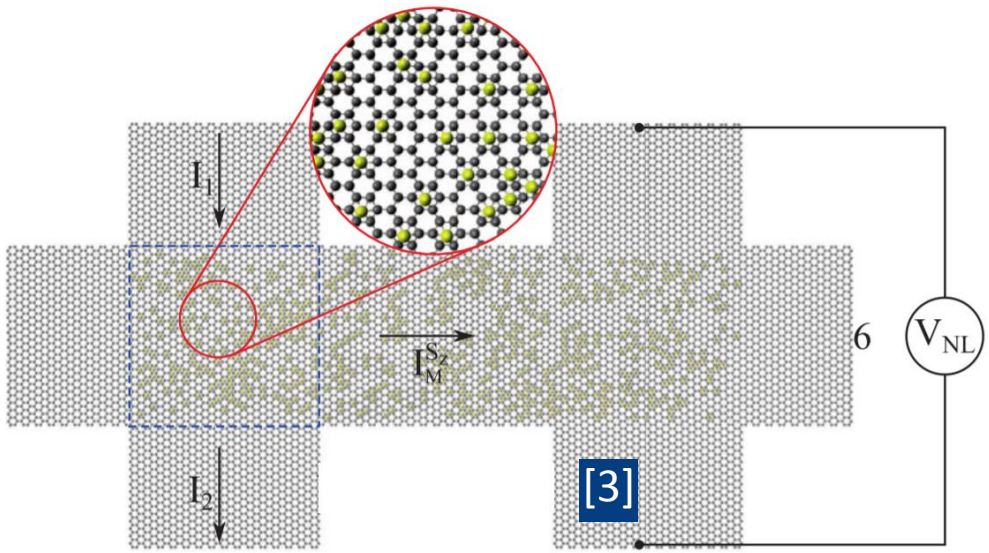
Graphene – 'the material of the millennium'



- **high sensitivity:**



- **high tunability:**

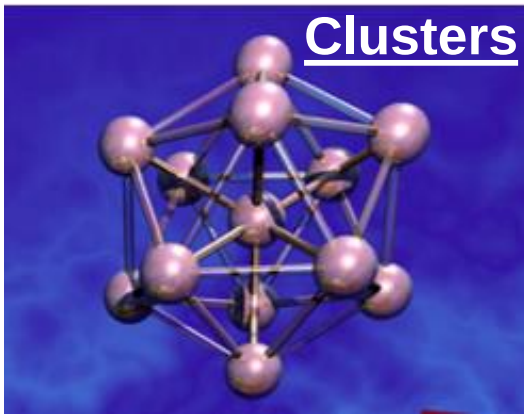


[2] Schedin *et al*, *Nature Mat.* **6**, 652 (2007)

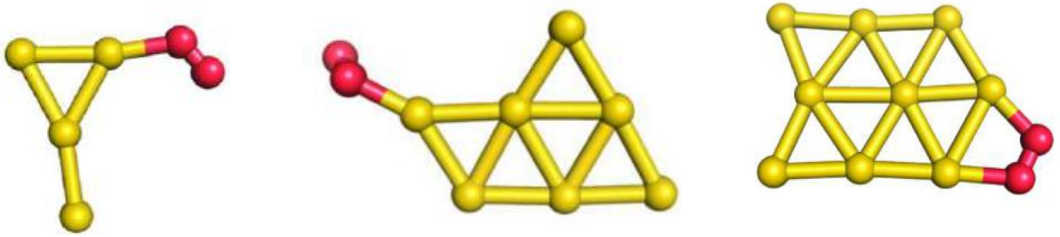
[3] D. Van Tuan *et al*, *PRL 117*, 176602 (2016)

The framework

Clusters – every atom counts

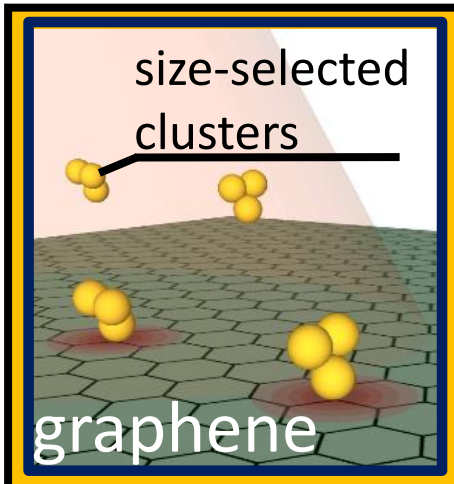


Au O [1]



-0.24 eV -0.53 eV -0.38 eV

$$CO + \frac{1}{2}O_2 \longleftrightarrow CO_2$$

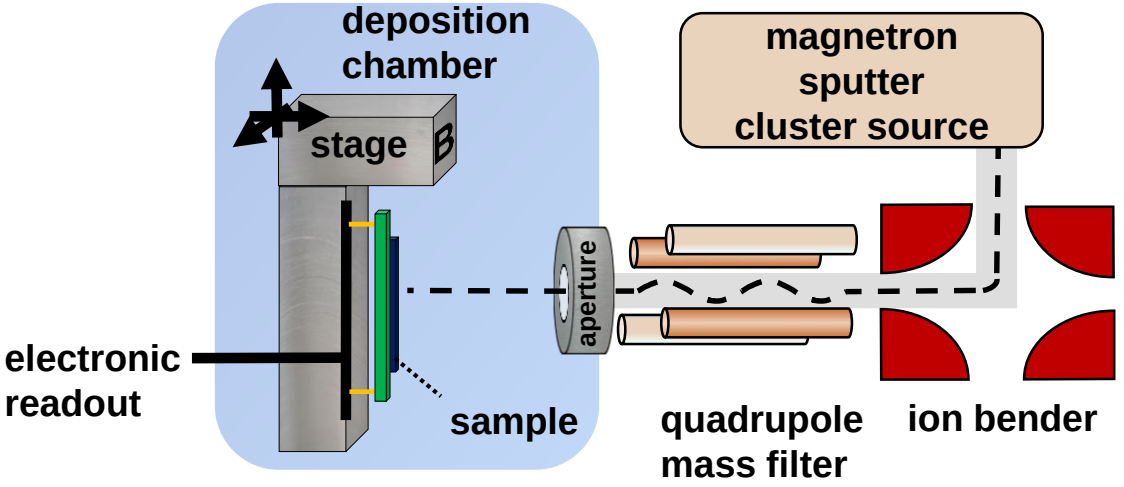


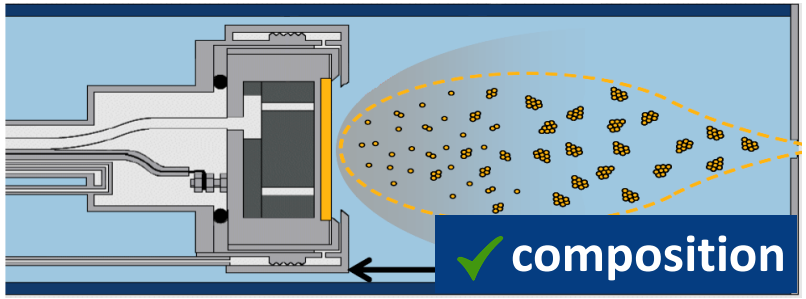
- **Graphene functionalization**
- **Cluster sensor**

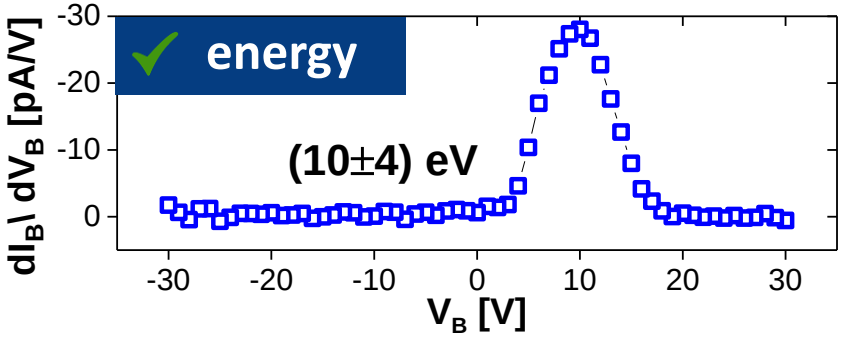
[1] A. Woodham et al,
J. Am. Chem. Soc. **135**, 1727 (2013)

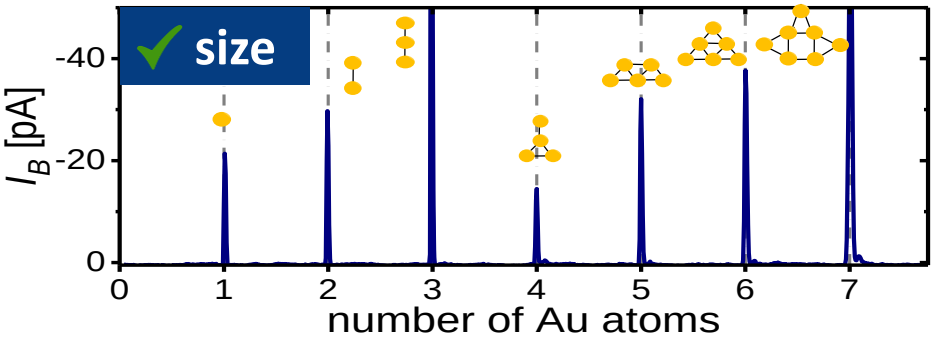
Cluster deposition

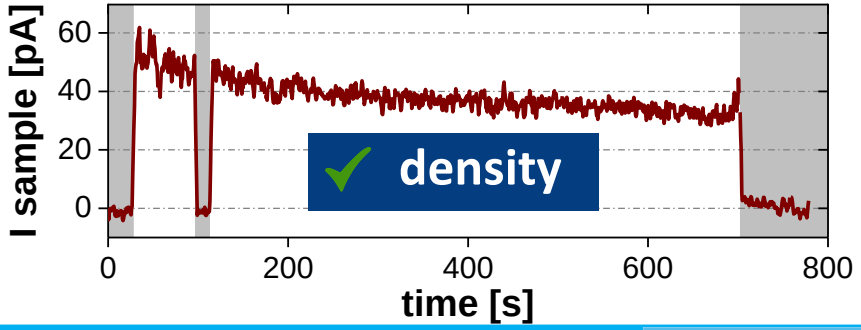
Ingredient I – High control





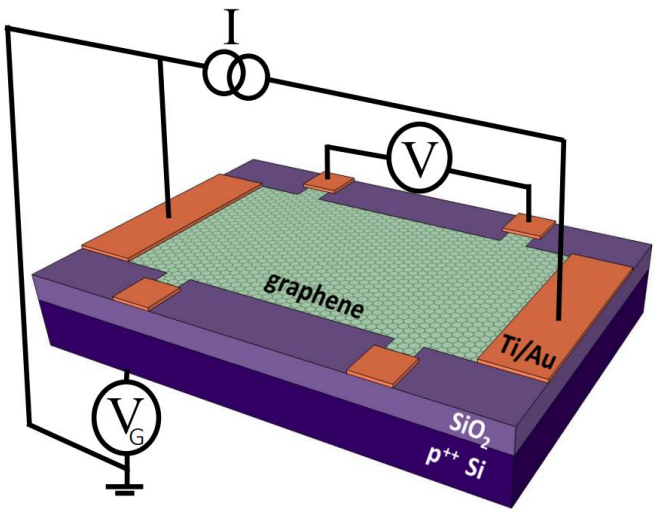


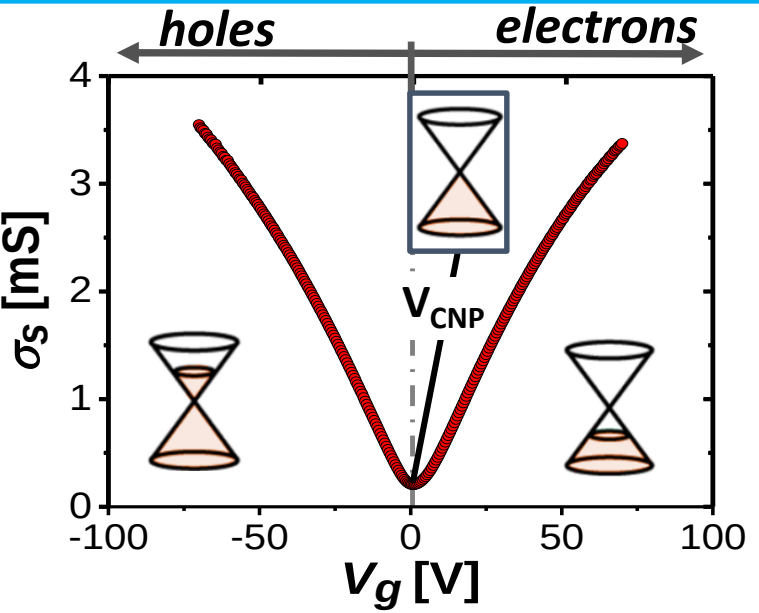


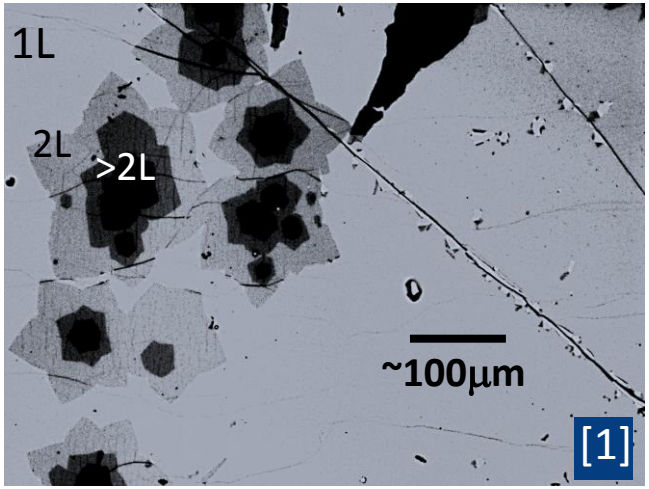


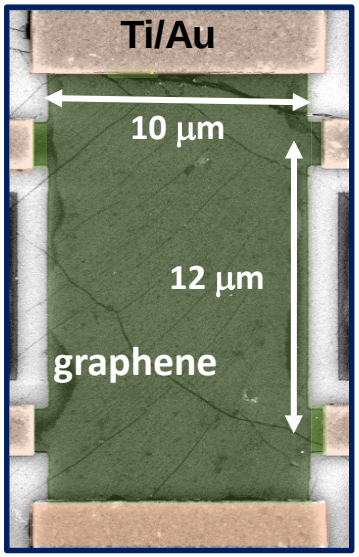
Graphene FET Devices

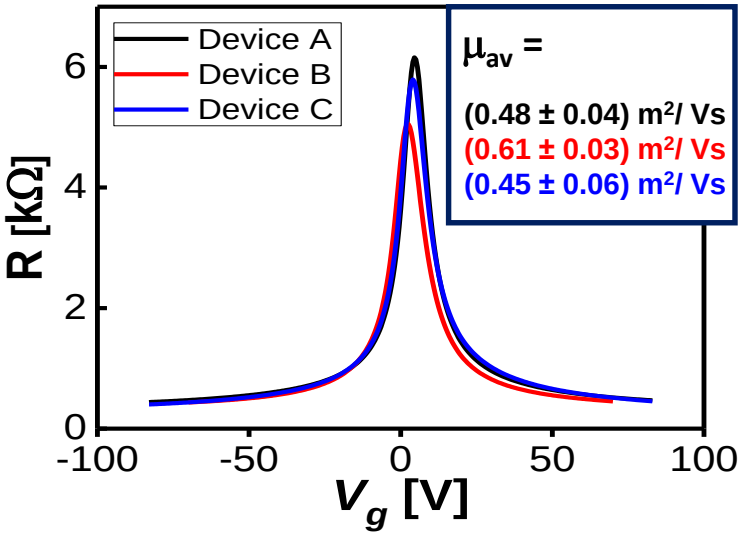
Ingredient II – field effect measurements







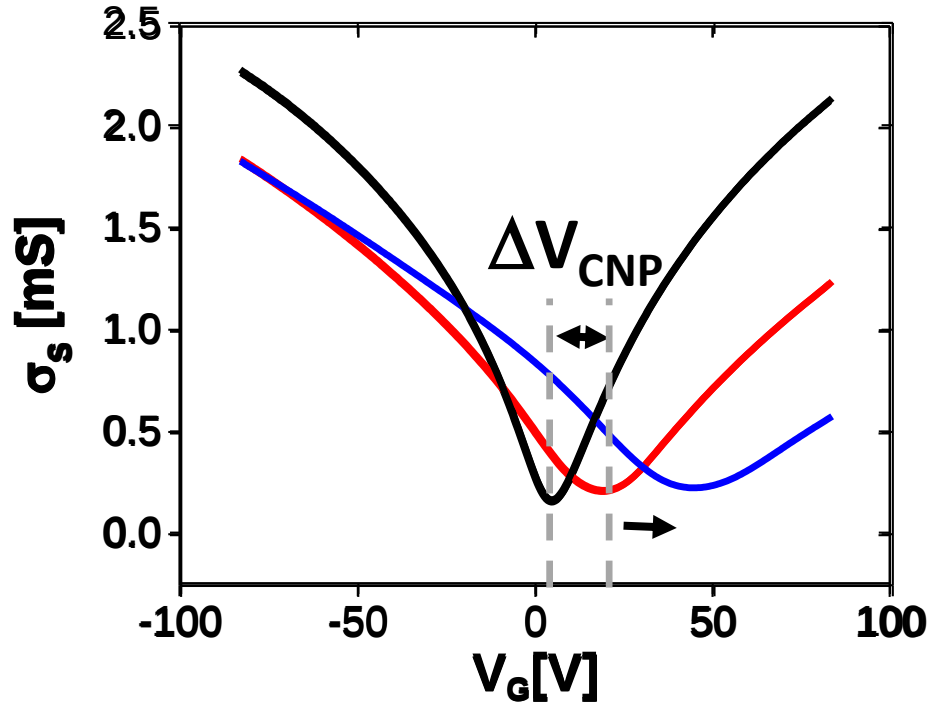


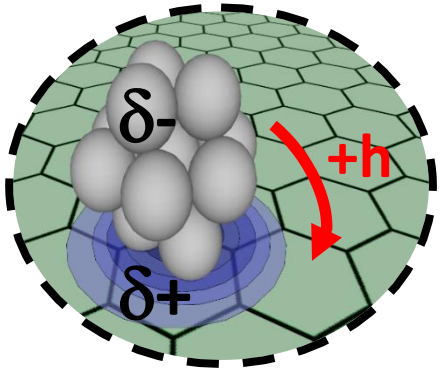


[1] N. Reckinger *et al*, Nanoscale **8**, 18751 (2016)

Clusters on graphene

Let's deposit!



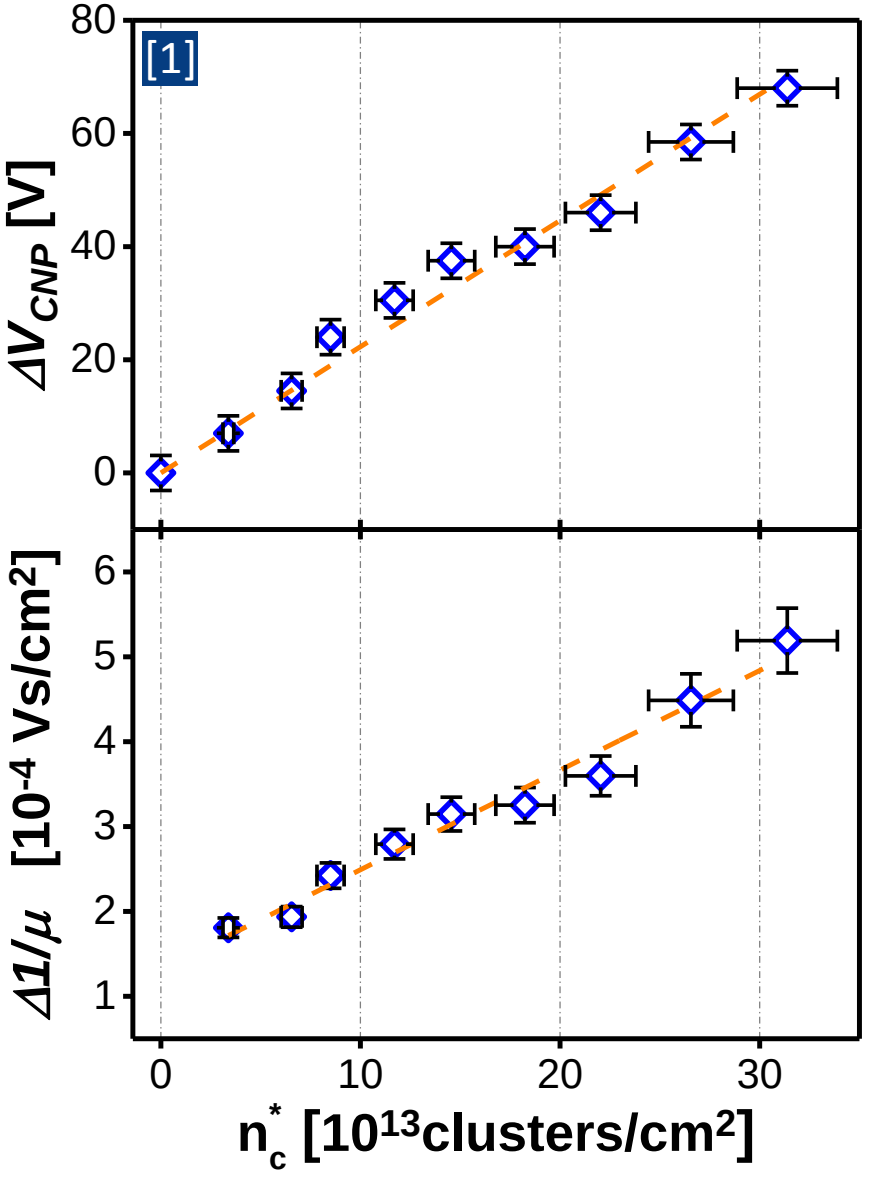


- **Linear trend**

$$\Delta V_{CNP} \propto \varepsilon \cdot n_c$$

$$1/\Delta\mu \propto n_c$$ [2]

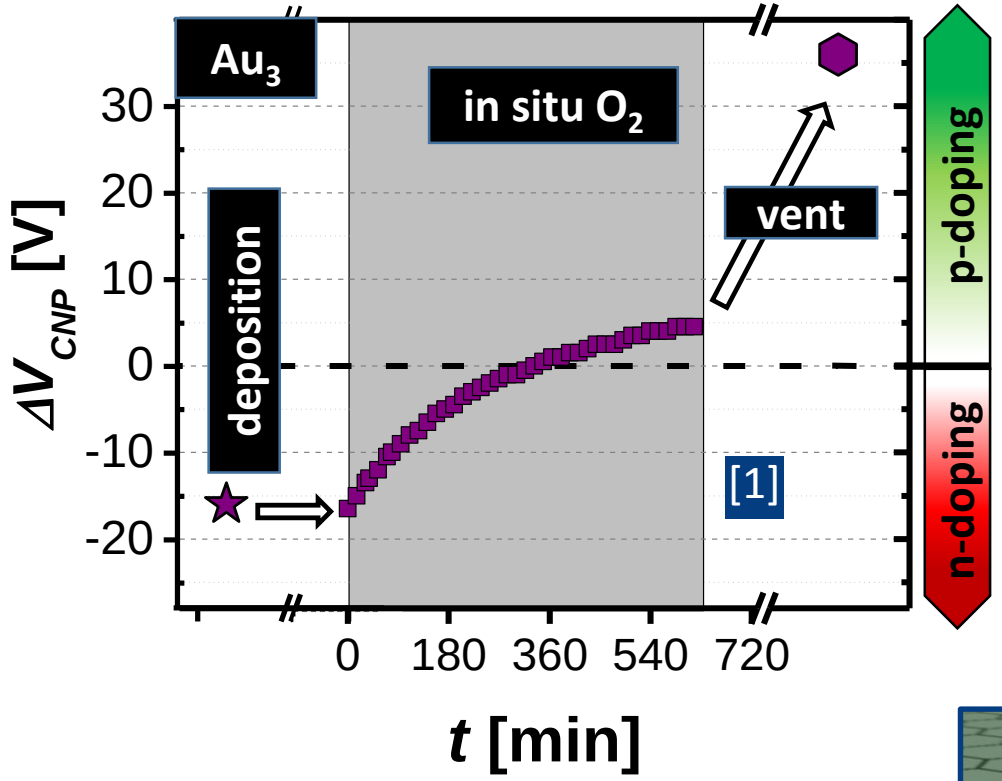
Charge donated per cluster

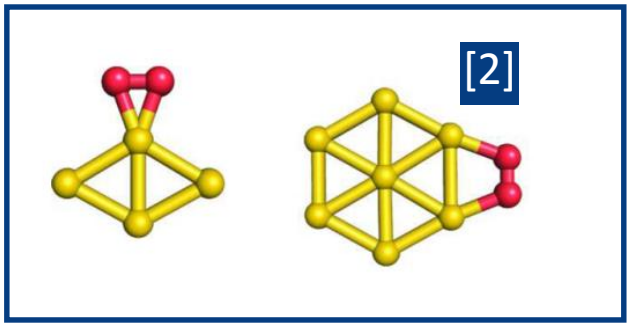


[1] J.E. Scheerder *et al*, Nanoscale 9, 10494 (2017) [2] S. Adam *et al*, PNAS 104, 18392 (2007)

Clusters on graphene

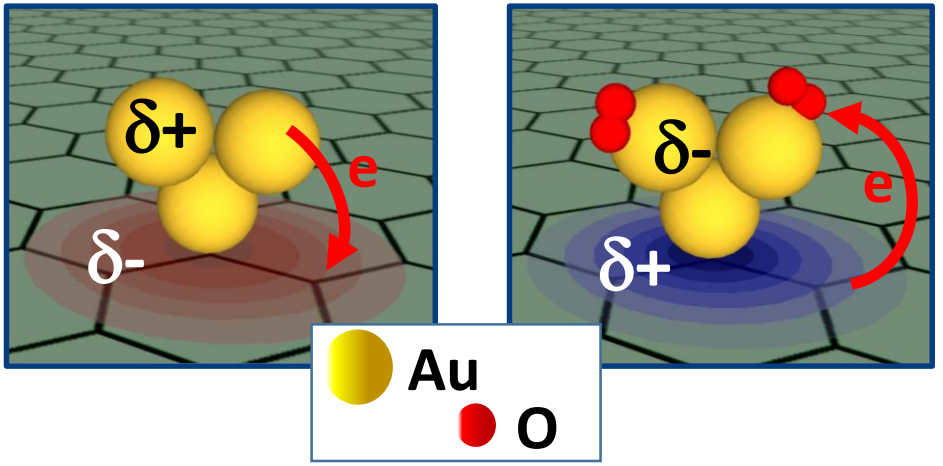
What can we do with it? - Catalysis





- **Graphene=**
  direct charge transfer probe

- **Oxygen adsorption to Au-cluster**
  = activation for catalysis!



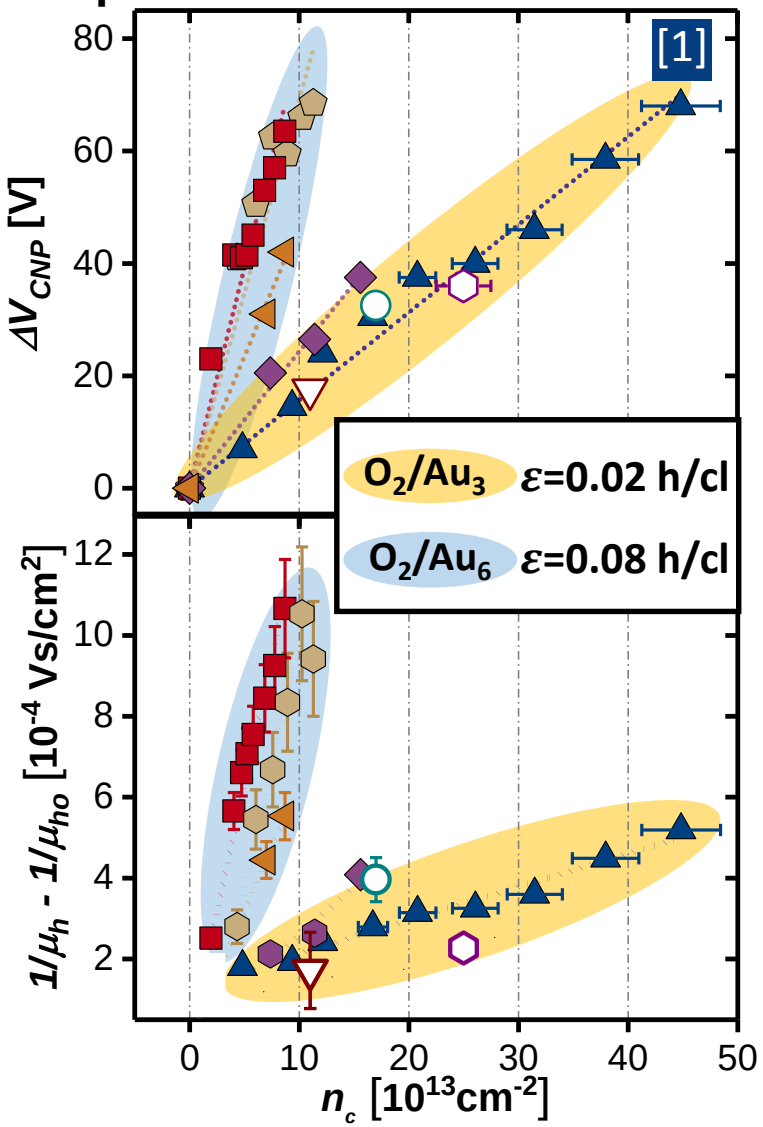
[1] J.E. Scheerder *et al*, submitted

[2] A. Woodham et al, J. Am. Chem. Soc. 135, 1727 (2013)

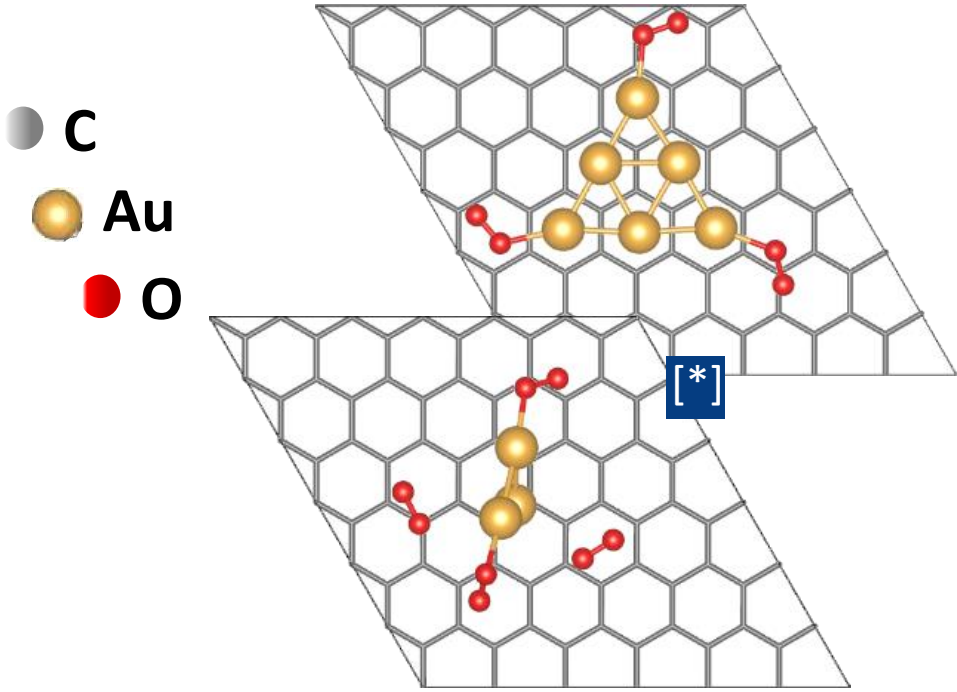
Clusters on graphene

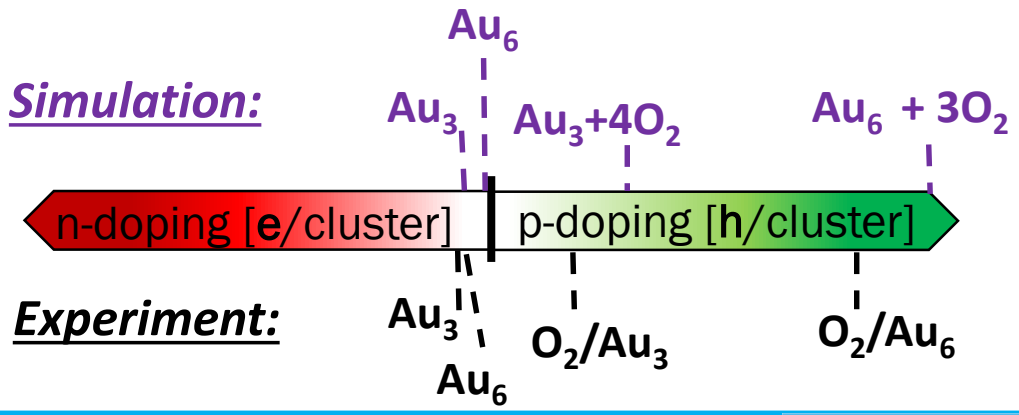
Size – effect !

Experiment:



DFT computation:





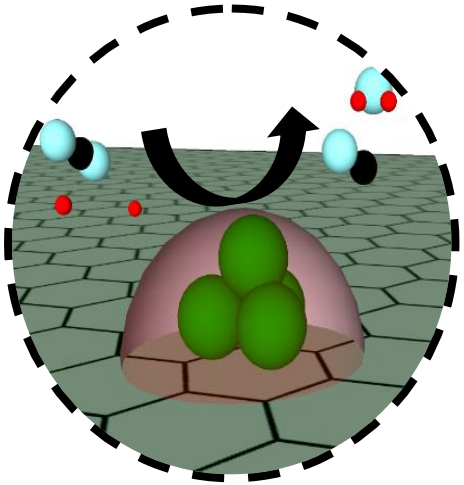
[1] J.E. Scheerder *et al*, submitted

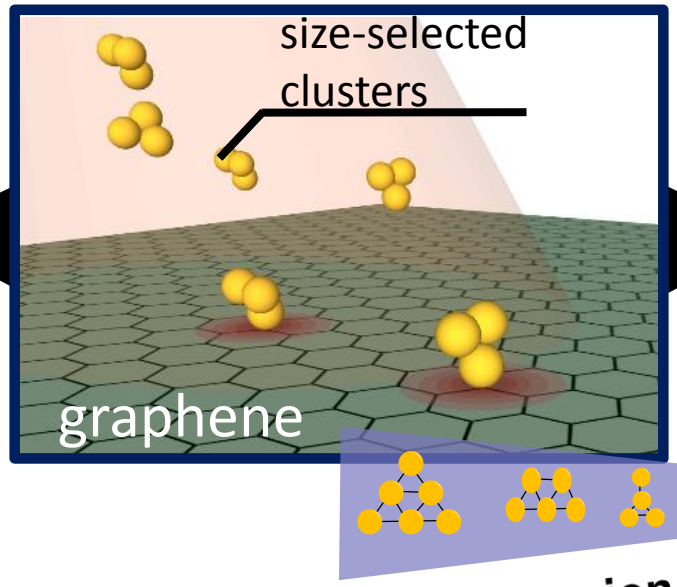
[*] thanks to prof. H.-P. Cheng, Dr. S. Liu (UFlorida)

Clusters on graphene

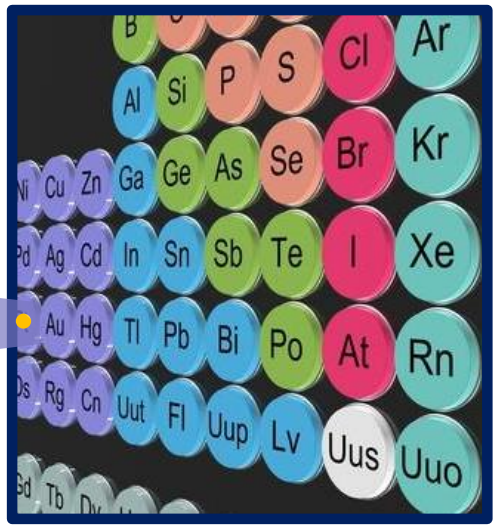
The scientific roadmap

Cluster sensor



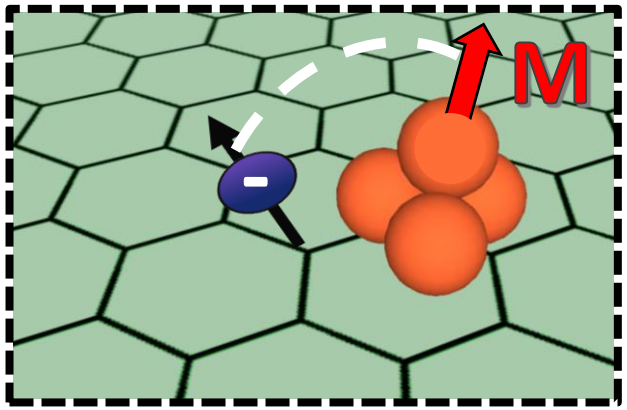


Graphene functionalization

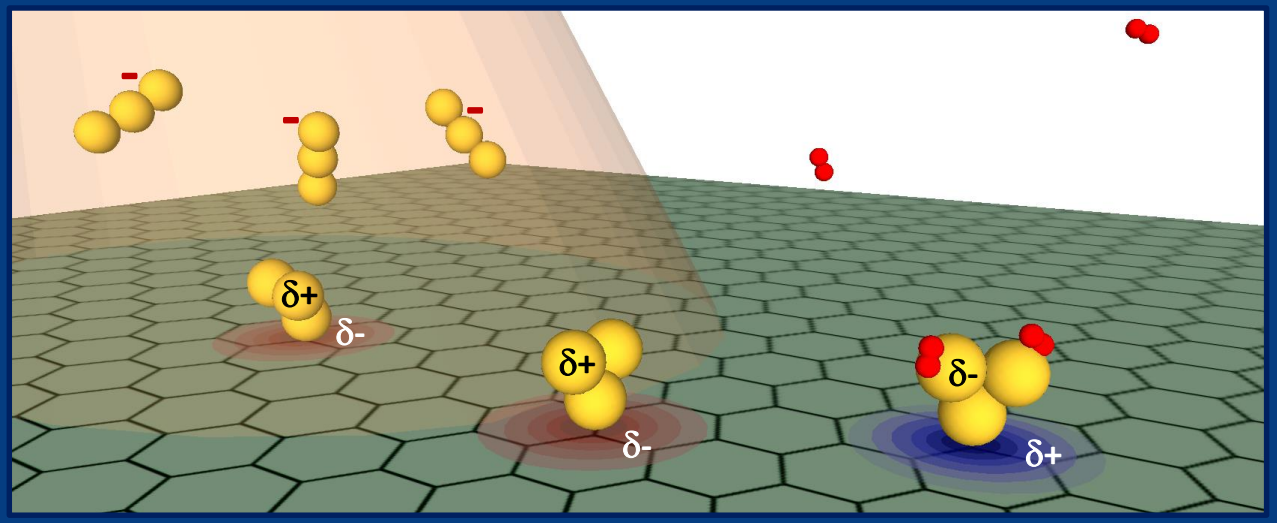


Catalysis with size-selected clusters:

"… [1,2] *…novel* ***characterization tools*** *to unravel mechanisms in* ***catalysis*** *and viable fabrication methods to* ***prepare particle decorated nanosheets****"*



[1] E. Tyo, S. Vajda, *Nat. Nanotech. 10, 577 (2015)* [2] D. Deng *et al*, *Nat. Nanotech. 11, 218 (2016)*



Thank you for attending !

KU LEUVEN
prof. J. Van de Vondel, prof. E. Janssens
Dr. Thomas Picot, V.S. Zharinov, W. Keijers,
Dr. B. Raes, R. Panghotra

CARBONNAGE UNIVERSITÉ DE NAMUR
prof. J.-F. Colomer, Dr. N. Reckinger

micas KU LEUVEN
prof. B. Puers, Dr. F. Ceyssens

UF UNIVERSITY of FLORIDA quantum theory project
prof. H.-P. Cheng, Dr. S. Liu

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KU LEUVEN
28th June 2018

APPENDIX

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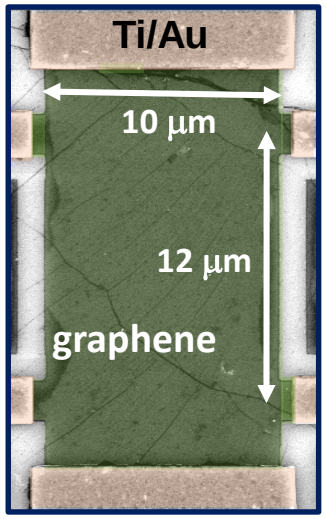
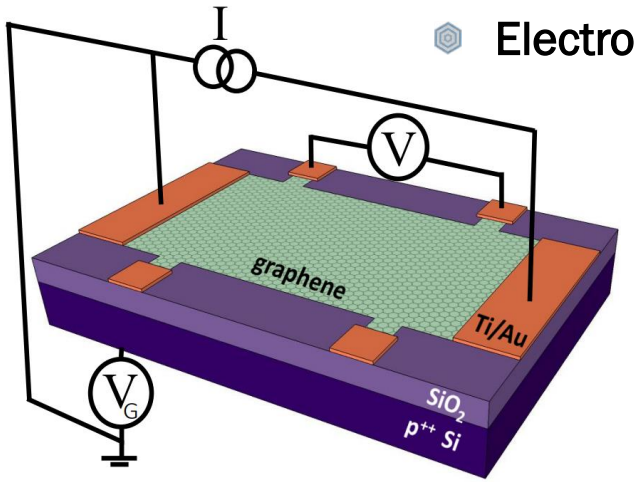
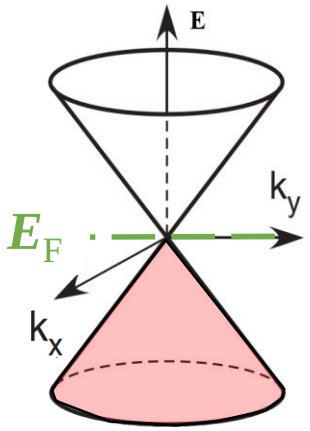
APPENDIX

FET and `Dirac' Cone

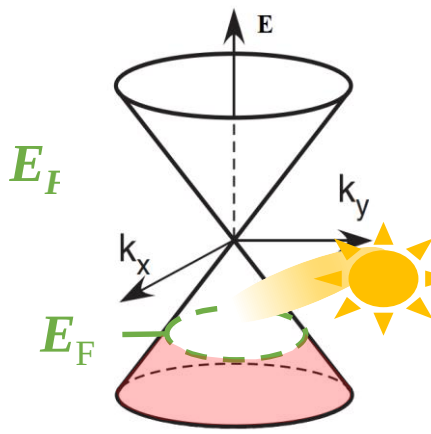




- Electronic structure graphene - gating and doping:



intrinsic



p-doped

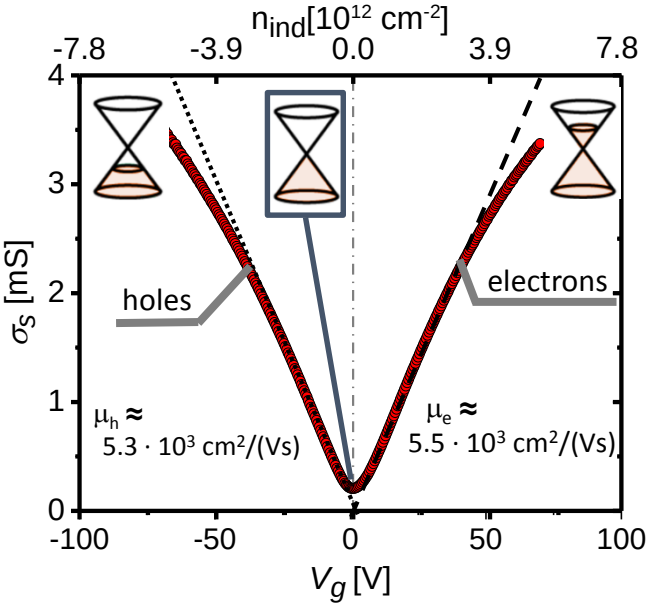
- **Field-effect:**

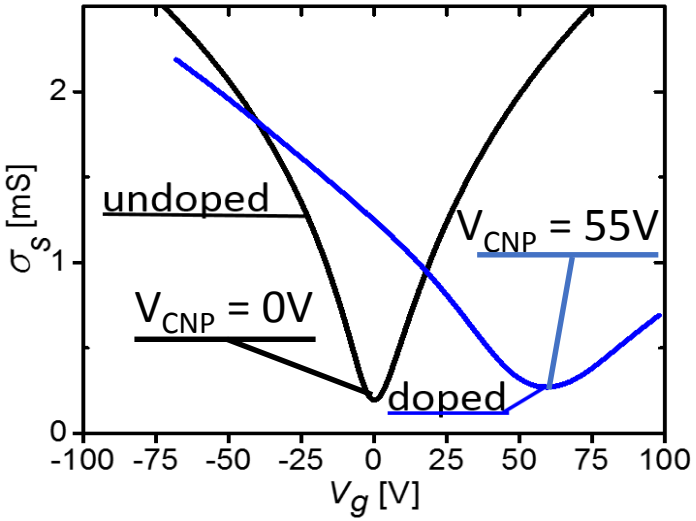
induced carriers:

$$n_{ind} = \alpha V_G$$

Mobility (Drude model)

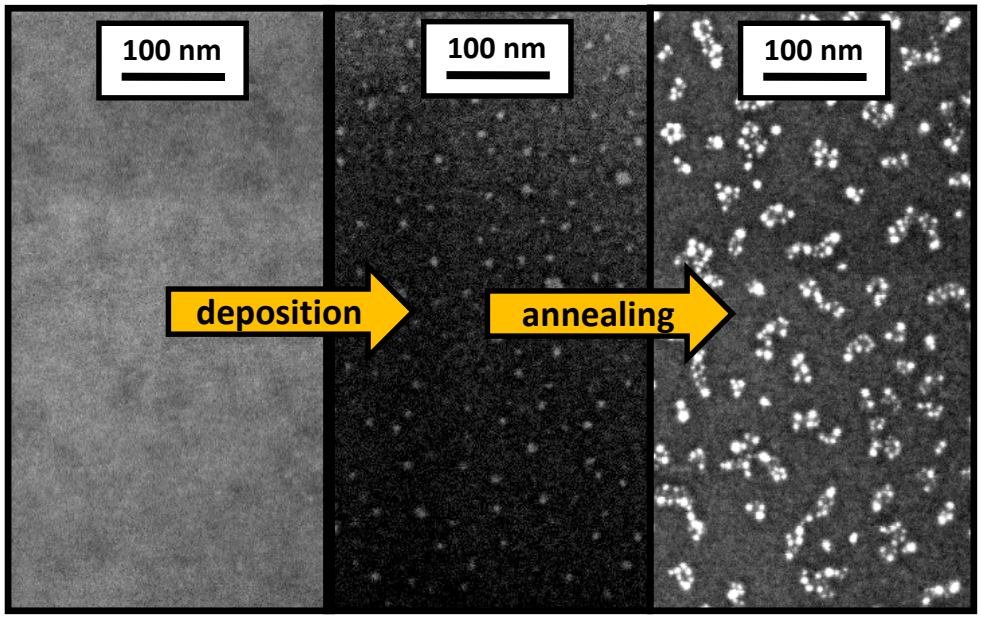
$$\sigma_s = n_{ind} e \mu$$



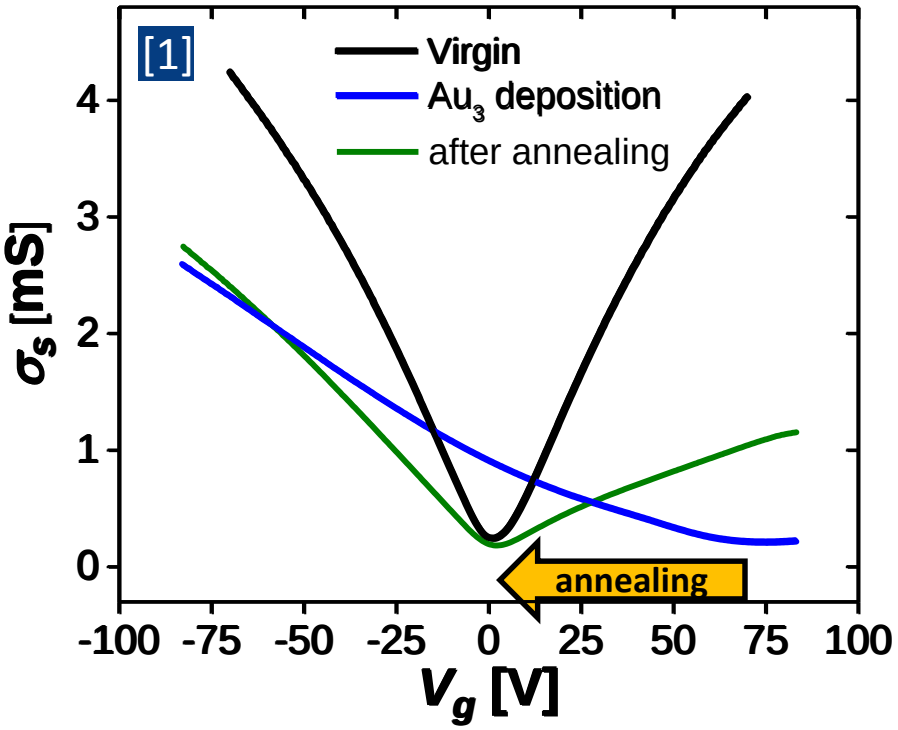


APPENDIX

how do they look like?



I ~ 3mA
P ~ 12mW
ΔT ~ Θ(100K)



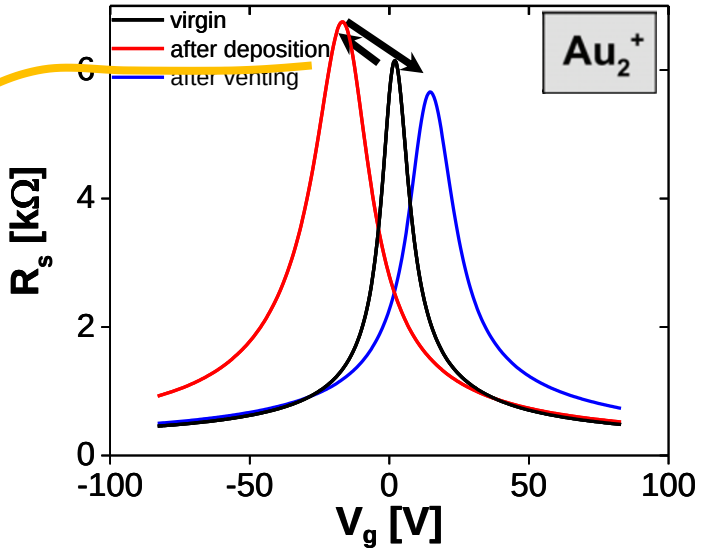
- Single clusters or sub-nm entities must be interacting !

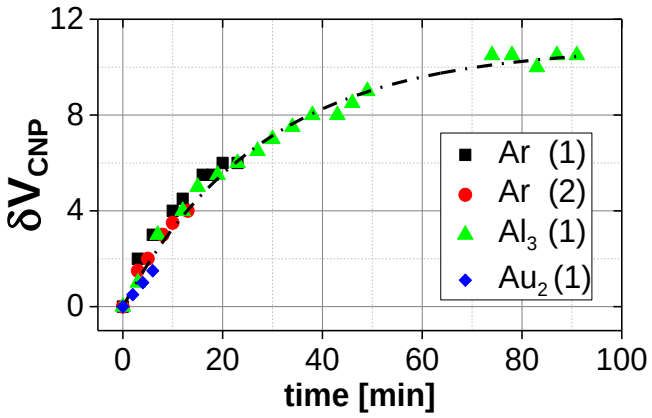
[1] J.E. Scheerder et al, Nanoscale 9, 10494 (2017)

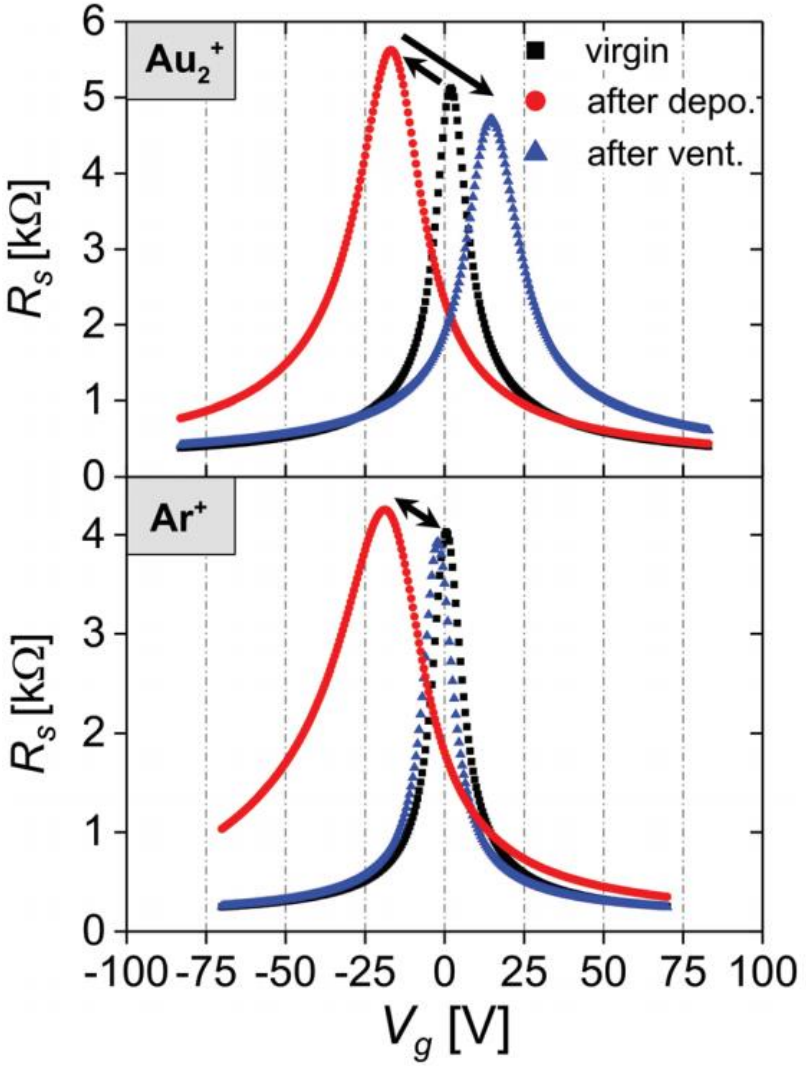
APPENDIX

Charging effect

unstable

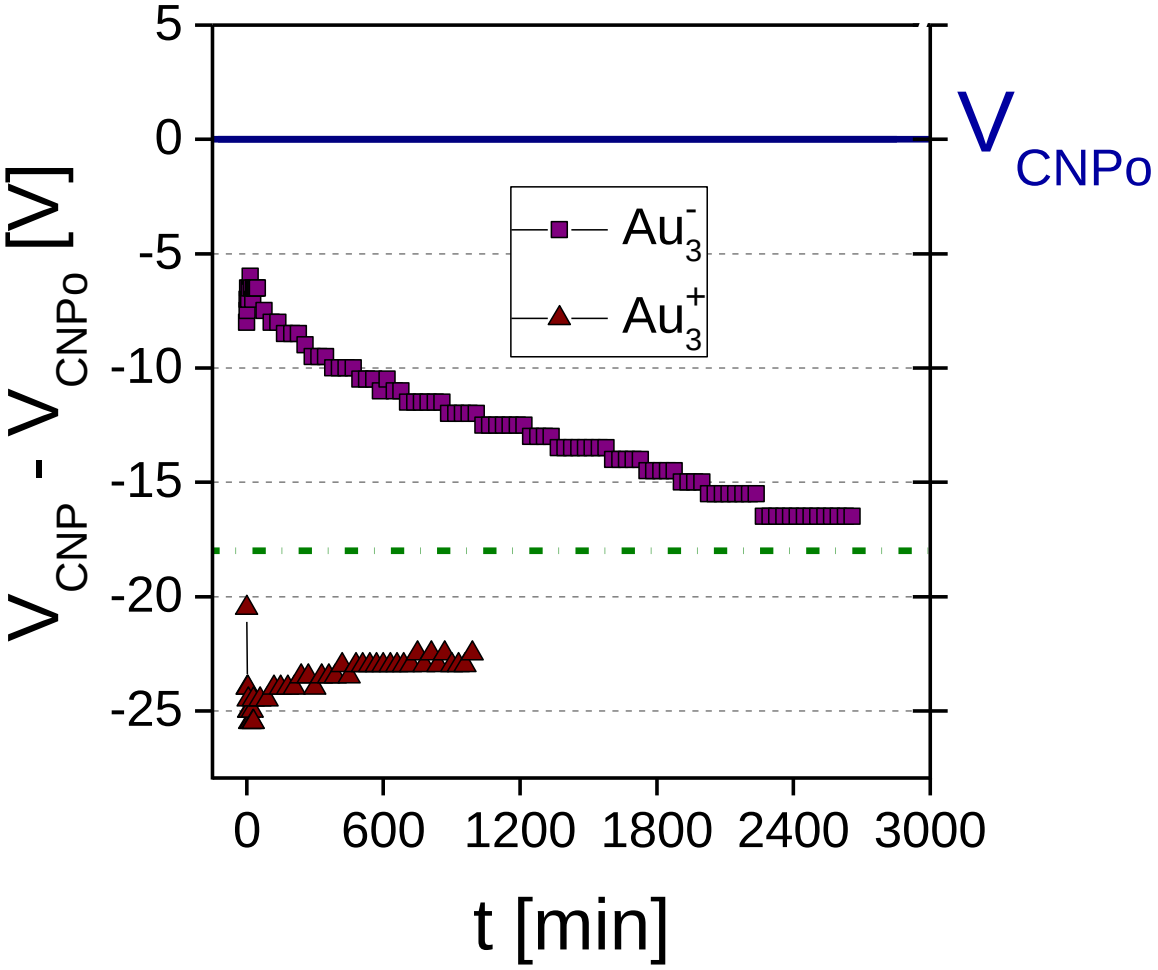


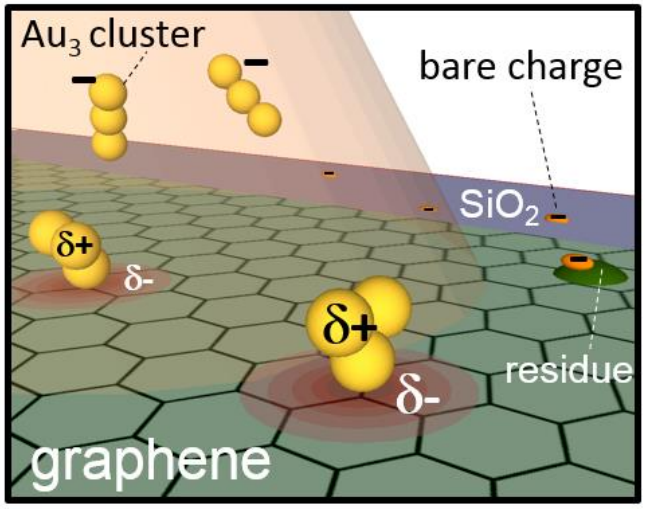


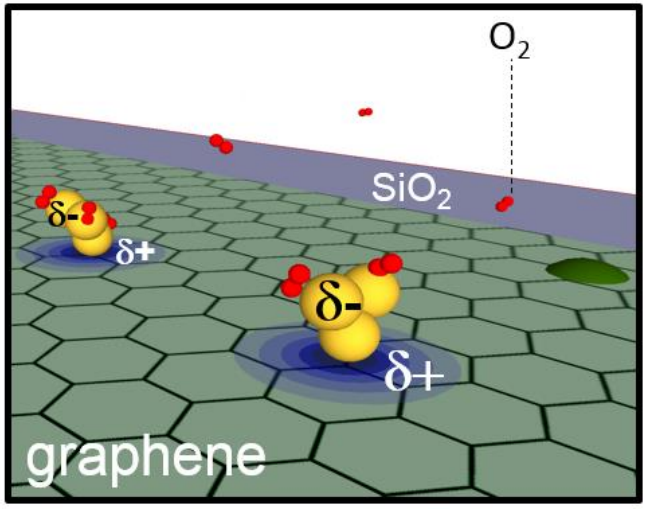


APPENDIX

Charging effect

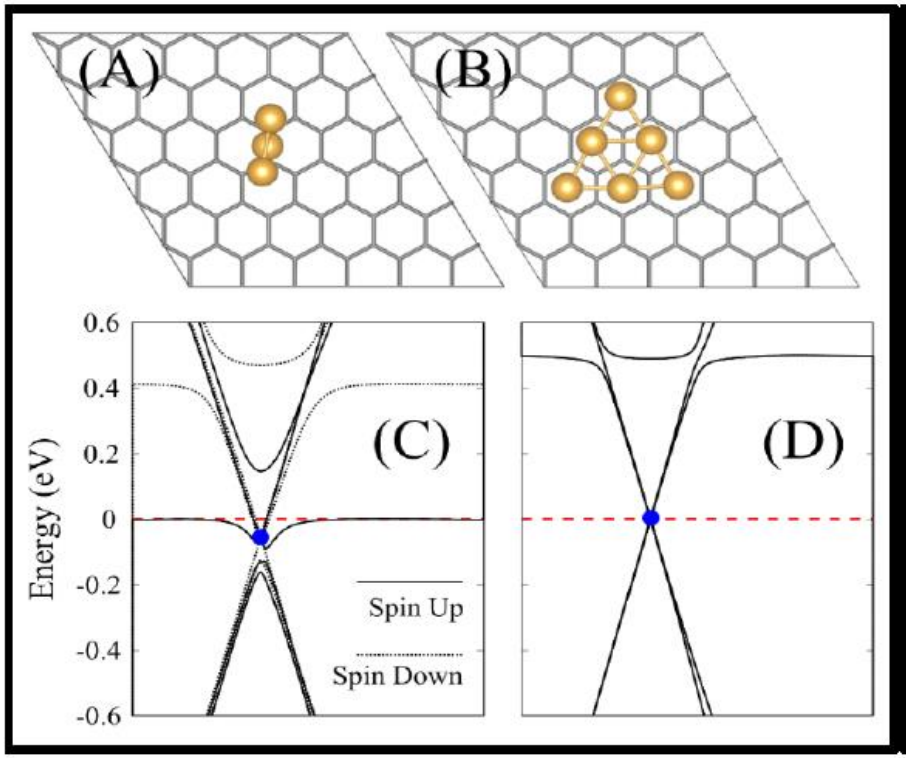


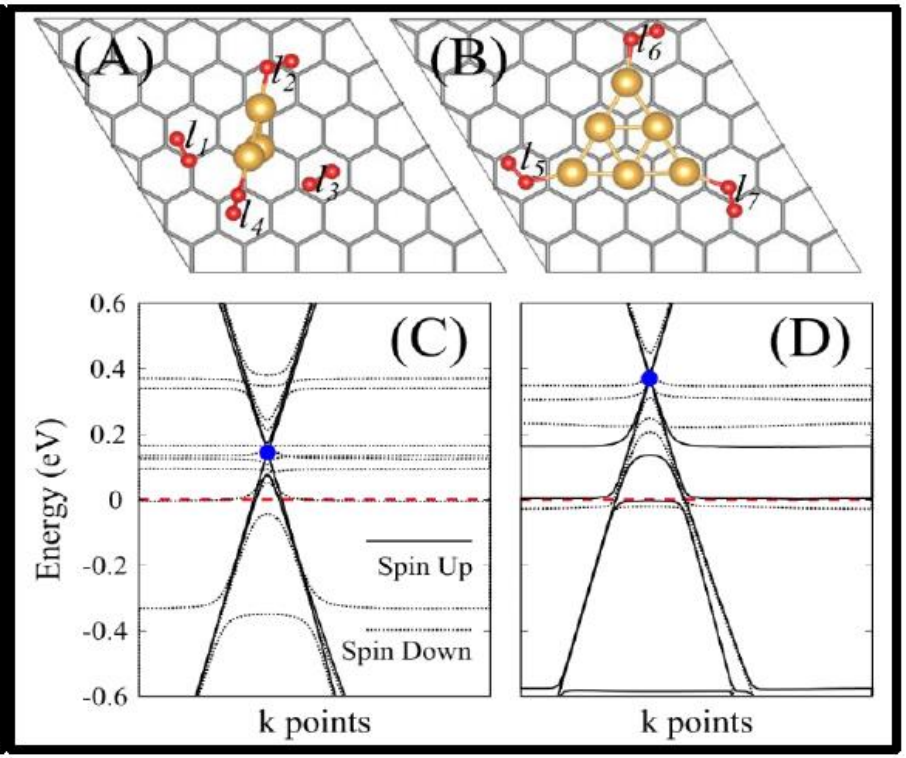


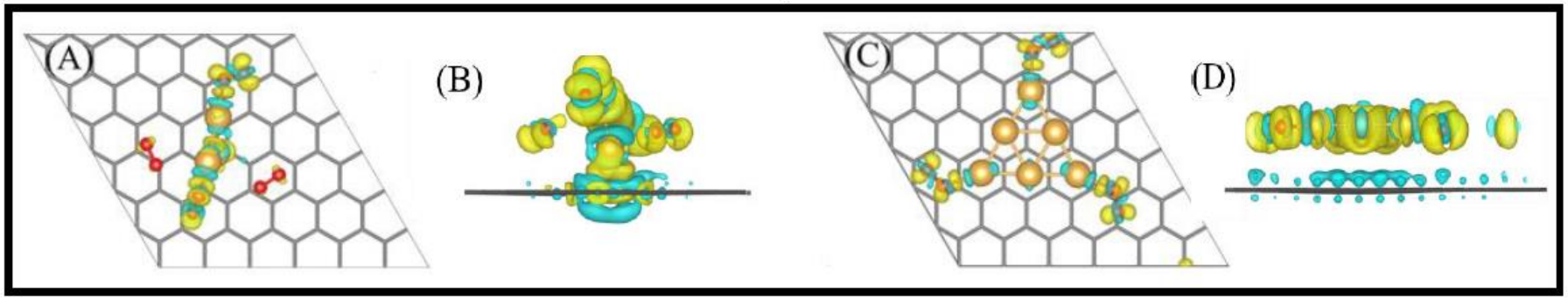


APPENDIX

DFT simulation

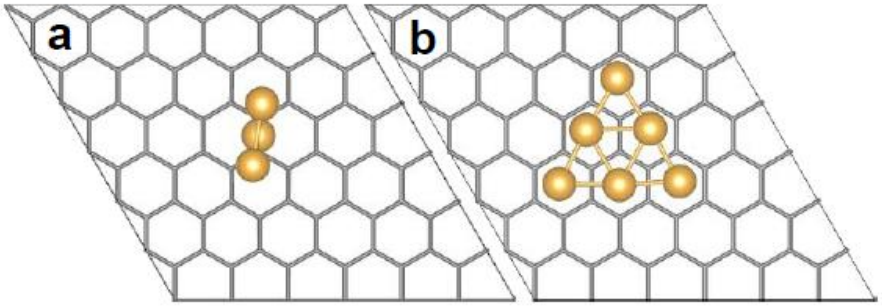


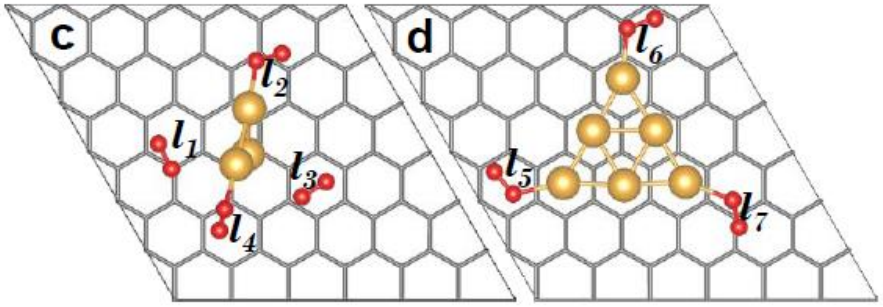


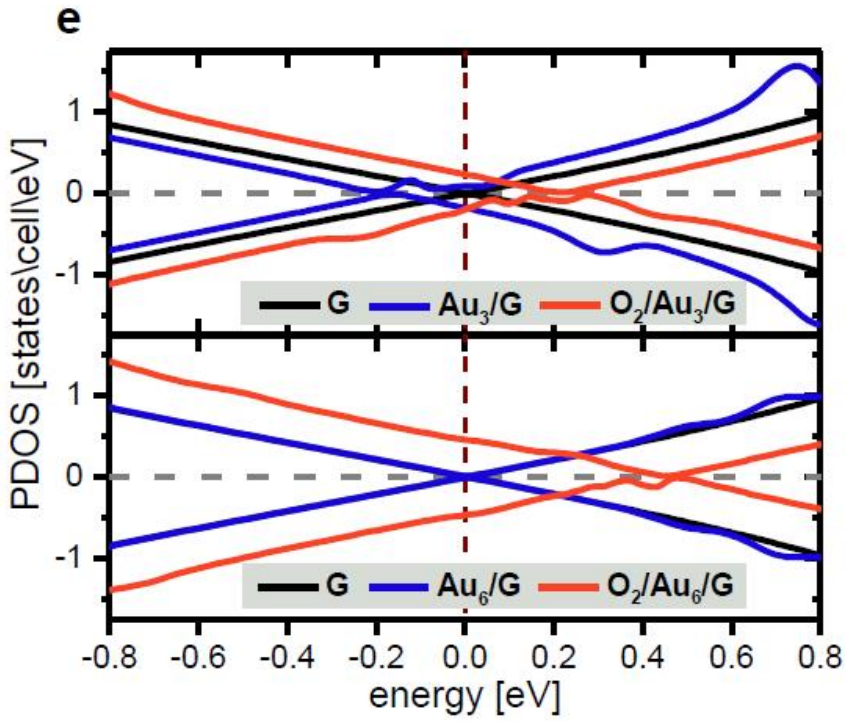


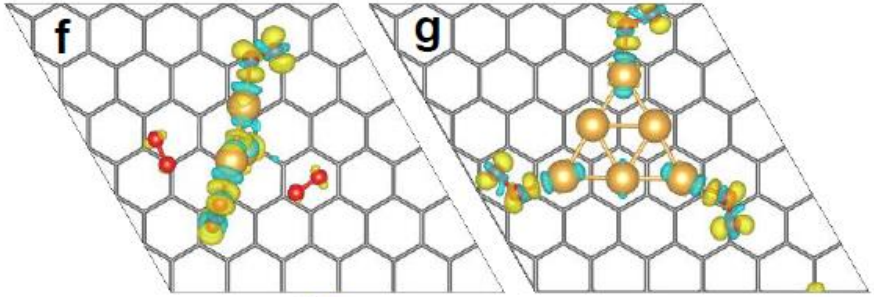
APPENDIX

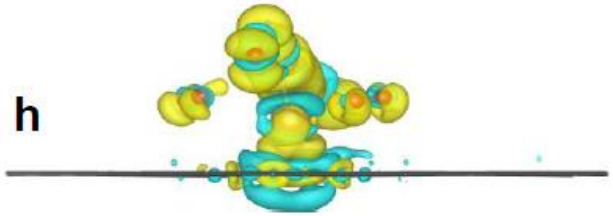
DFT simulation

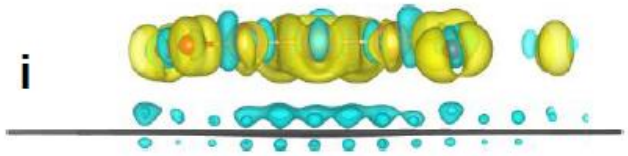






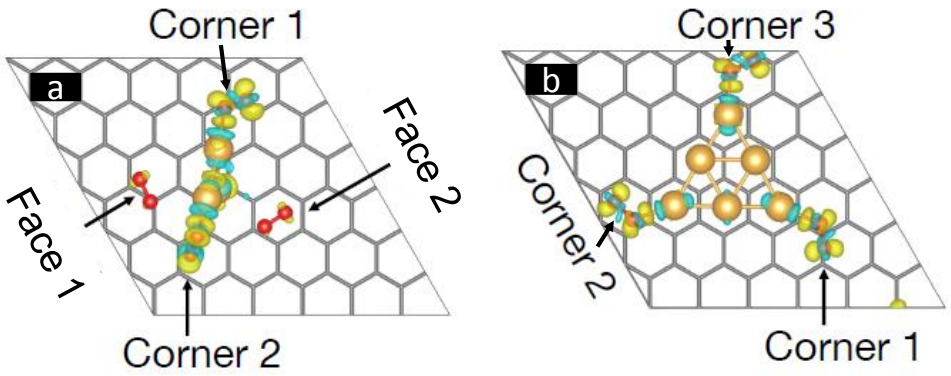






APPENDIX

DFT simulation

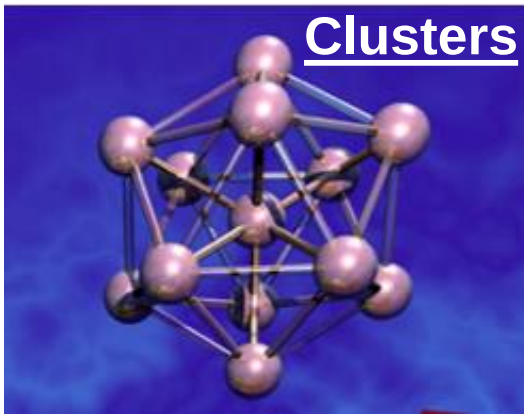


	Graphene	Au_3	O_2			
			Corner 1	Corner 2	Face 1	Face 2
B. charge transfer [*e*]	-0.269	-0.504	0.331	0.280	0.078	0.085
	-0.773		0.773			

	Graphene	Au6	O_2		
			Corner 1	Corner 2	Corner 3
B. charge transfer [*e*]	-0.337	-0.403	0.252	0.254	0.233
	-0.740		0.740		

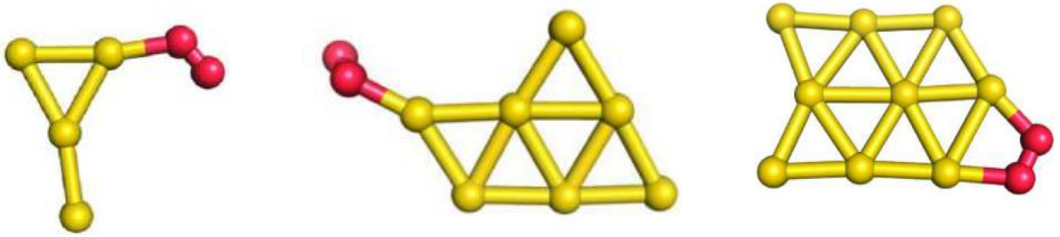
The framework

Clusters – every atom counts



[1]

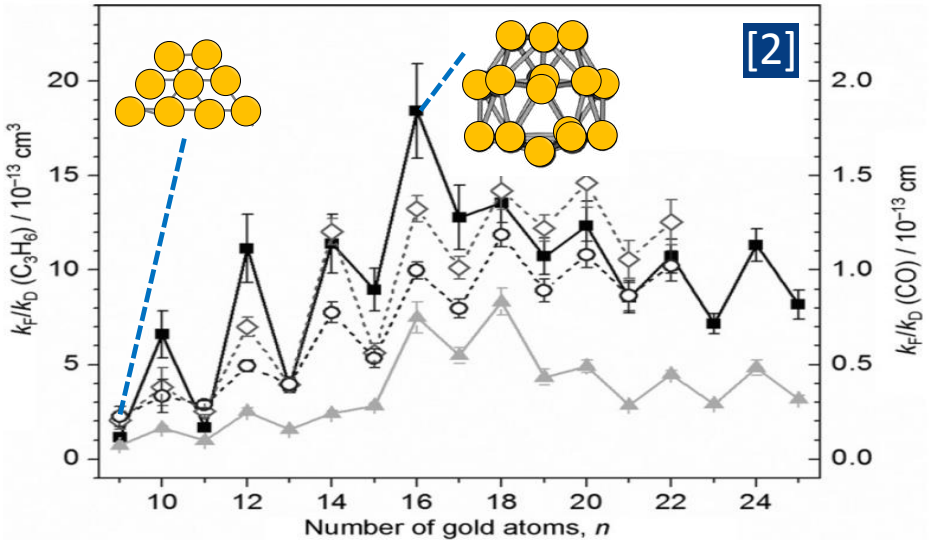
Au O



-0.24 eV -0.53 eV -0.38 eV

$$CO + \frac{1}{2} O_2 \longleftrightarrow CO_2$$

[2]



[1] A. Woodham et al,
J. Am. Chem. Soc. **135**, 1727 (2013)

[2] E. Janssens, P. Lievens et al,
Chem. Eu. J. **21**, 15256 (2015)