Multiscale modeling of 2D nanostructures for electronic and energy-related applications

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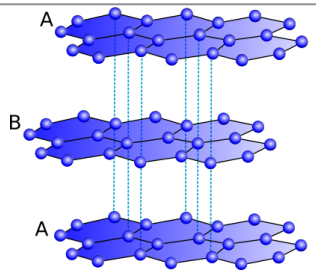
APS March Meeting, March 18-22, 2013

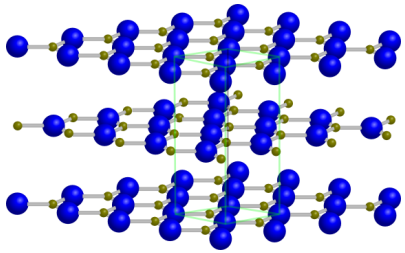
Baltimore MD

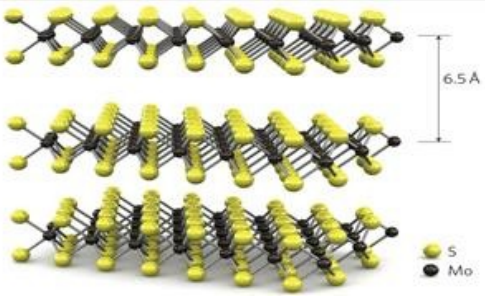
Graphene-multilayers

h-BN

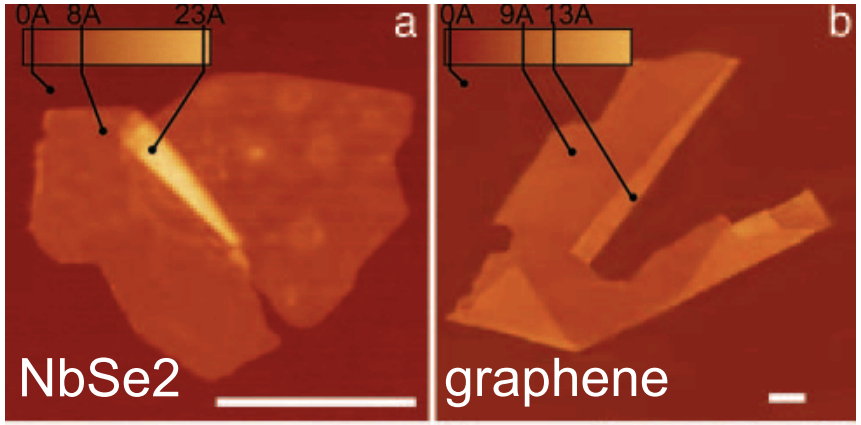
Metal di-chalcogenide
(WS_2, TiS_2, ZrS_2, $MoSe_2$, MoS_2, ...)

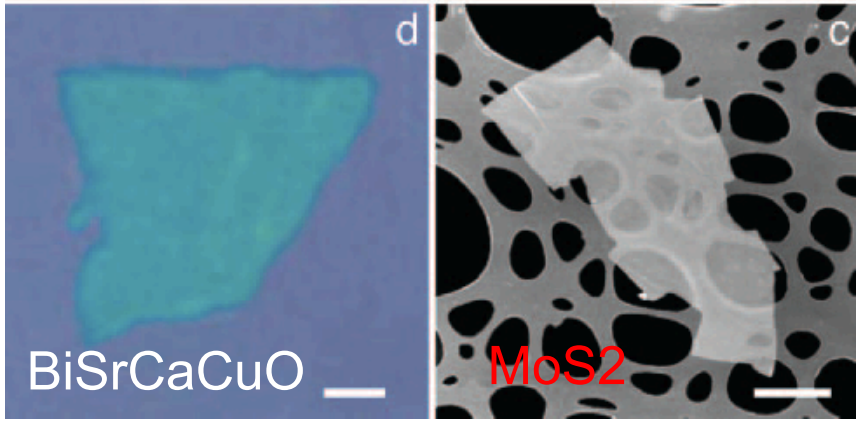


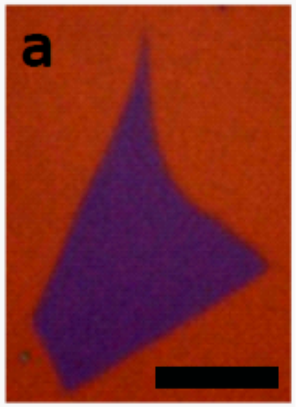


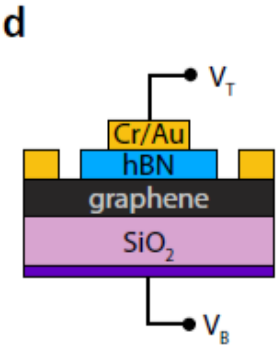


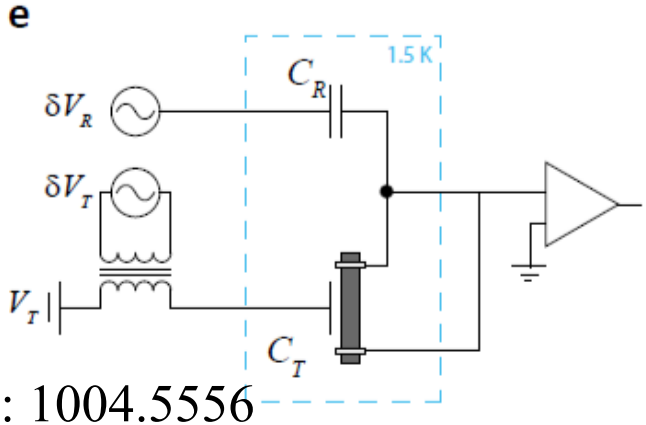
E_{gap} 0.0 eV 5.0 eV 1.7 eV











PNAS, **102**, 10451 (2005)

arXiv: 1004.5556

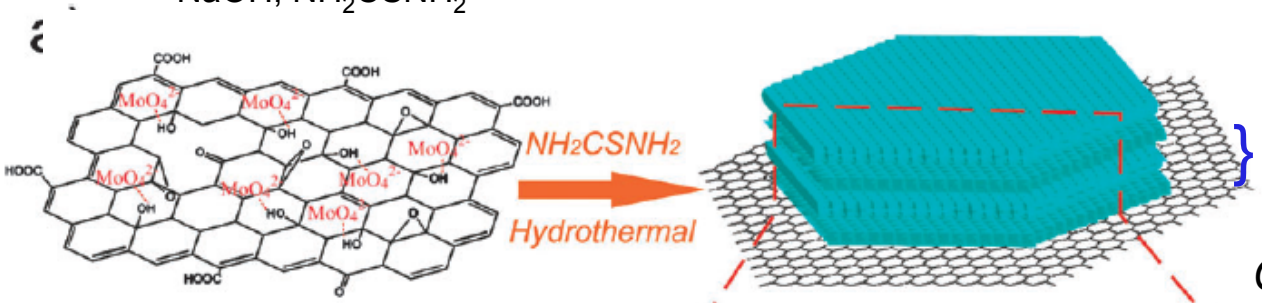
Graphene-MoS_2 interface: Experiments

Starting materials

Sodium molybdate ($Na_2MoO_4.2H_2O$),
graphene oxide (GO)
NaOH, NH_2CSNH_2

Products

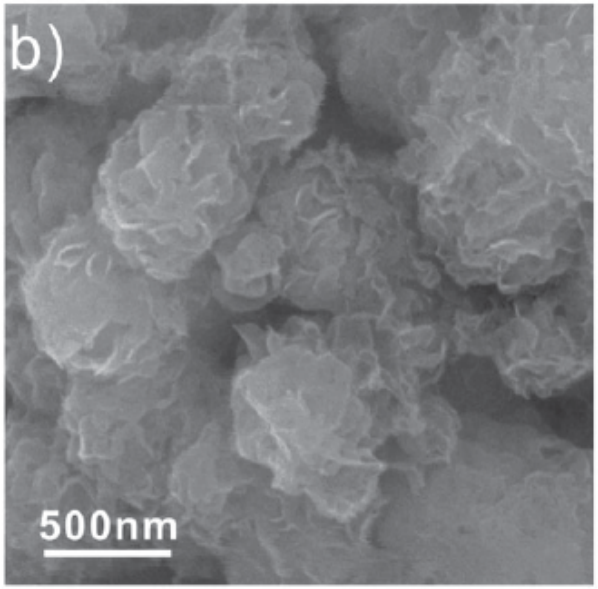
G/MoS_2 composites



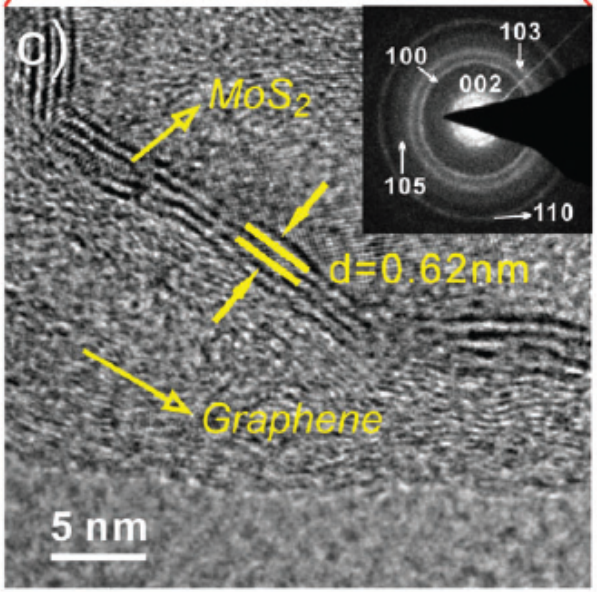
} 4-5 layers

Chem. Commun. (2011), **47**, 4252
ACS Nano, (2011), **5**, 4720

SEM (G/MoS_2 composites)

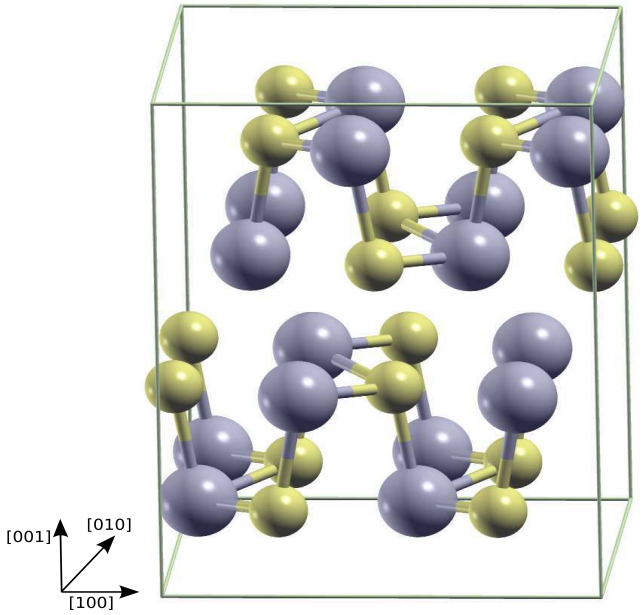


HRTEM



SnS / GeS thin films for PV applications:

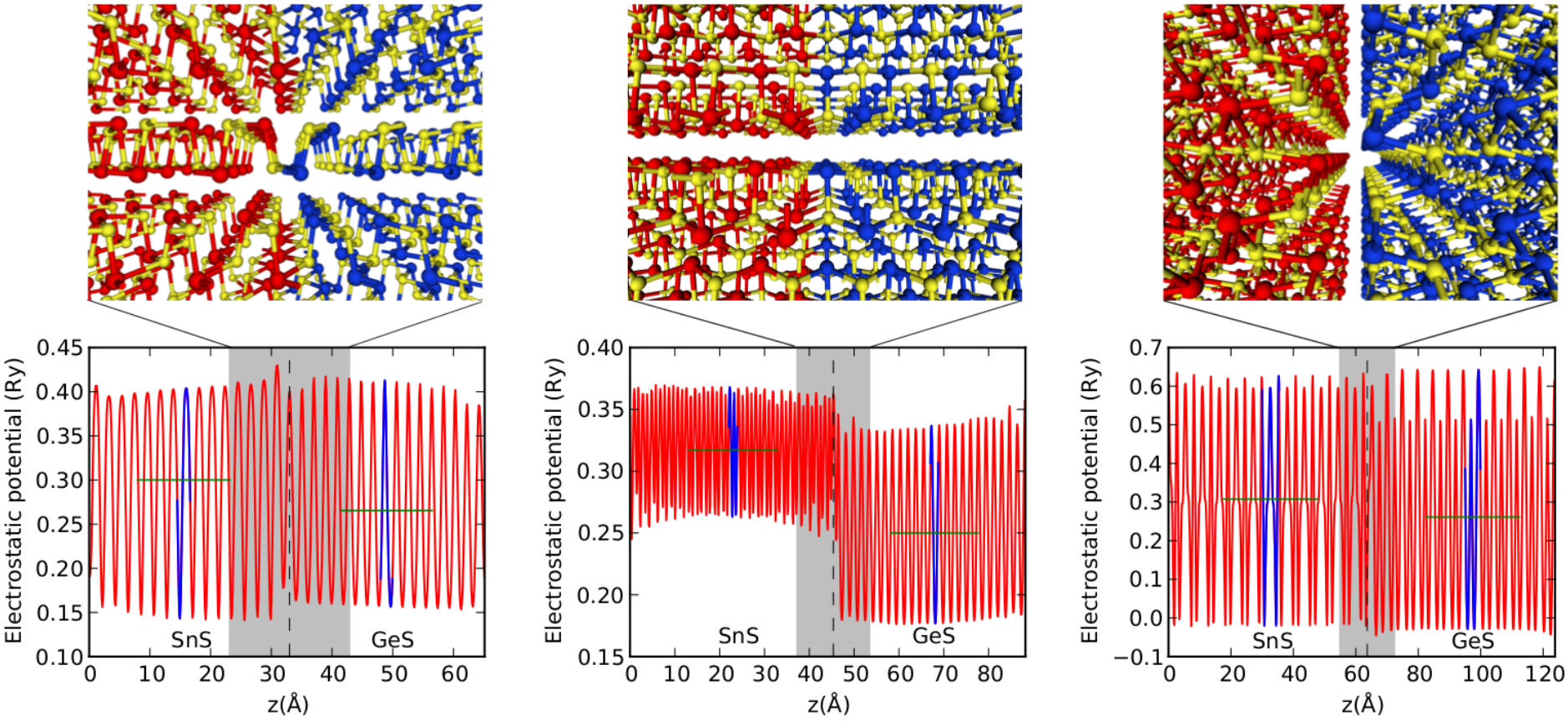
- high absorption ($\alpha > 10^4\ cm^{-1}$)
- suitable band gap (~1.1 – 1.5 eV)
- low-toxicity
- earth–abundant



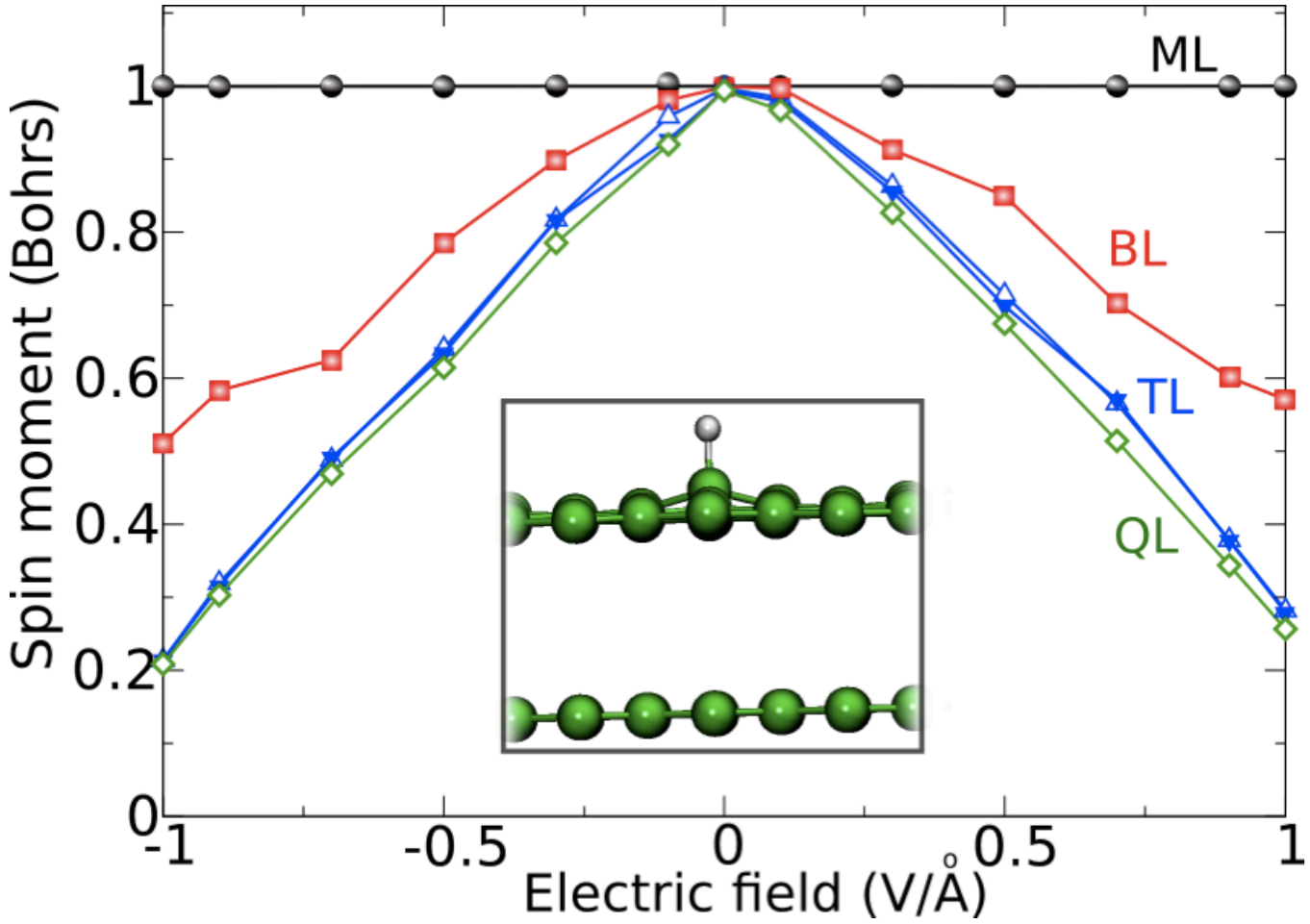
PV devices with efficiency up to ~2%
Roy Gordon group
Dept. of Chemistry - Harvard

See talk by Brad Malone on Thur. March 21, U23.00009

GeS/SnS interfaces for electron-hole separation – GW calculations

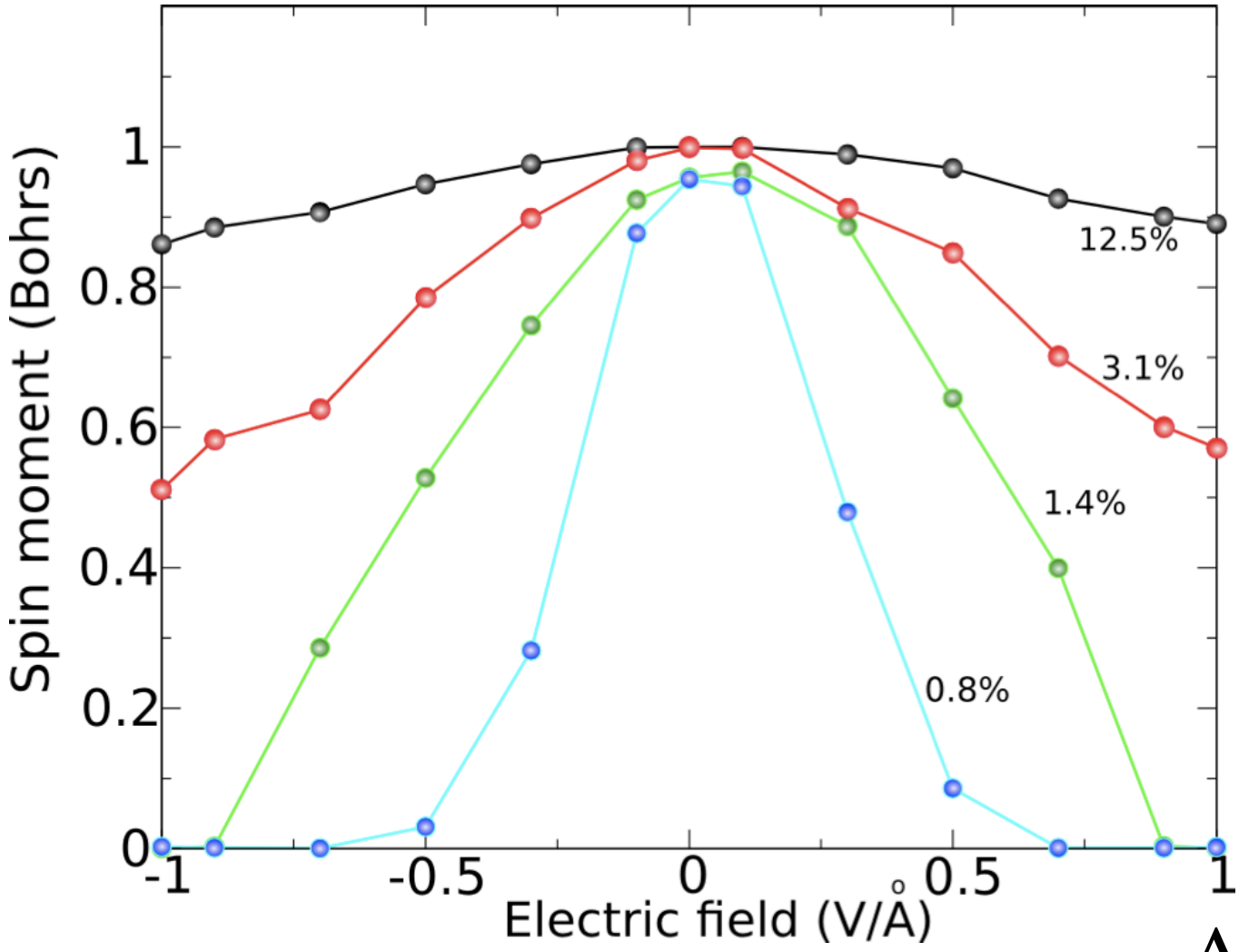


Magneto-electric effect in covalently functionalized few-layer graphene



concentration: ~3%

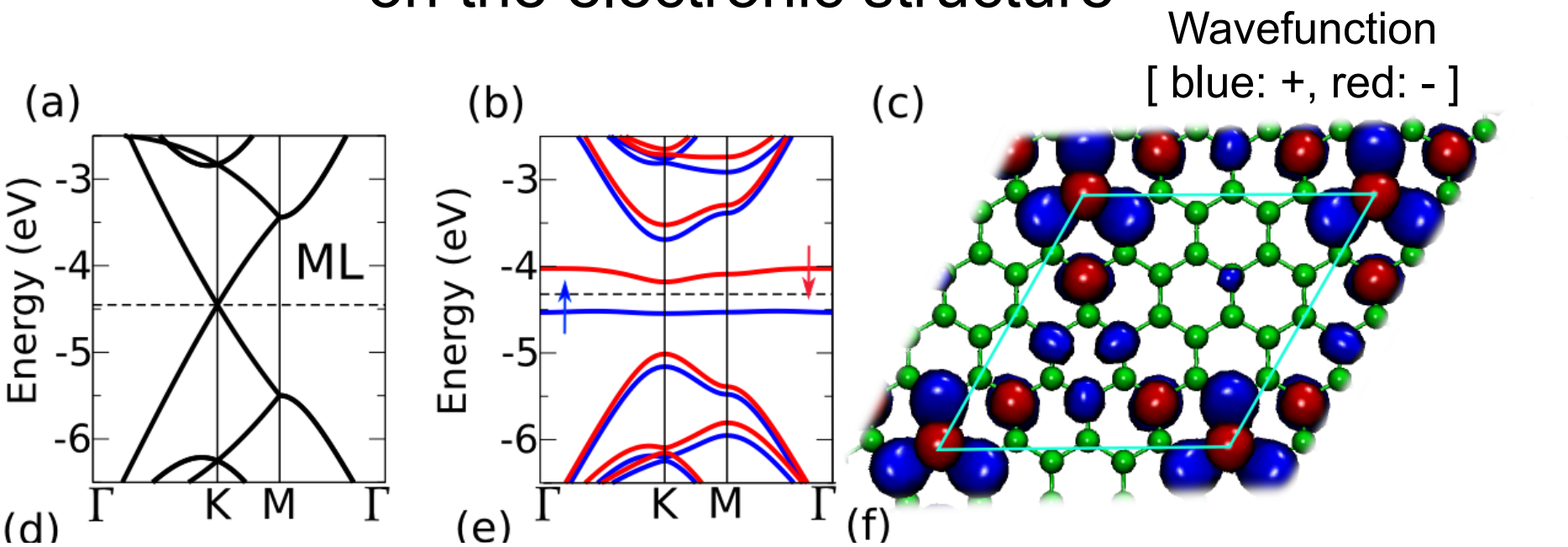
Impurity concentration plays a significant role



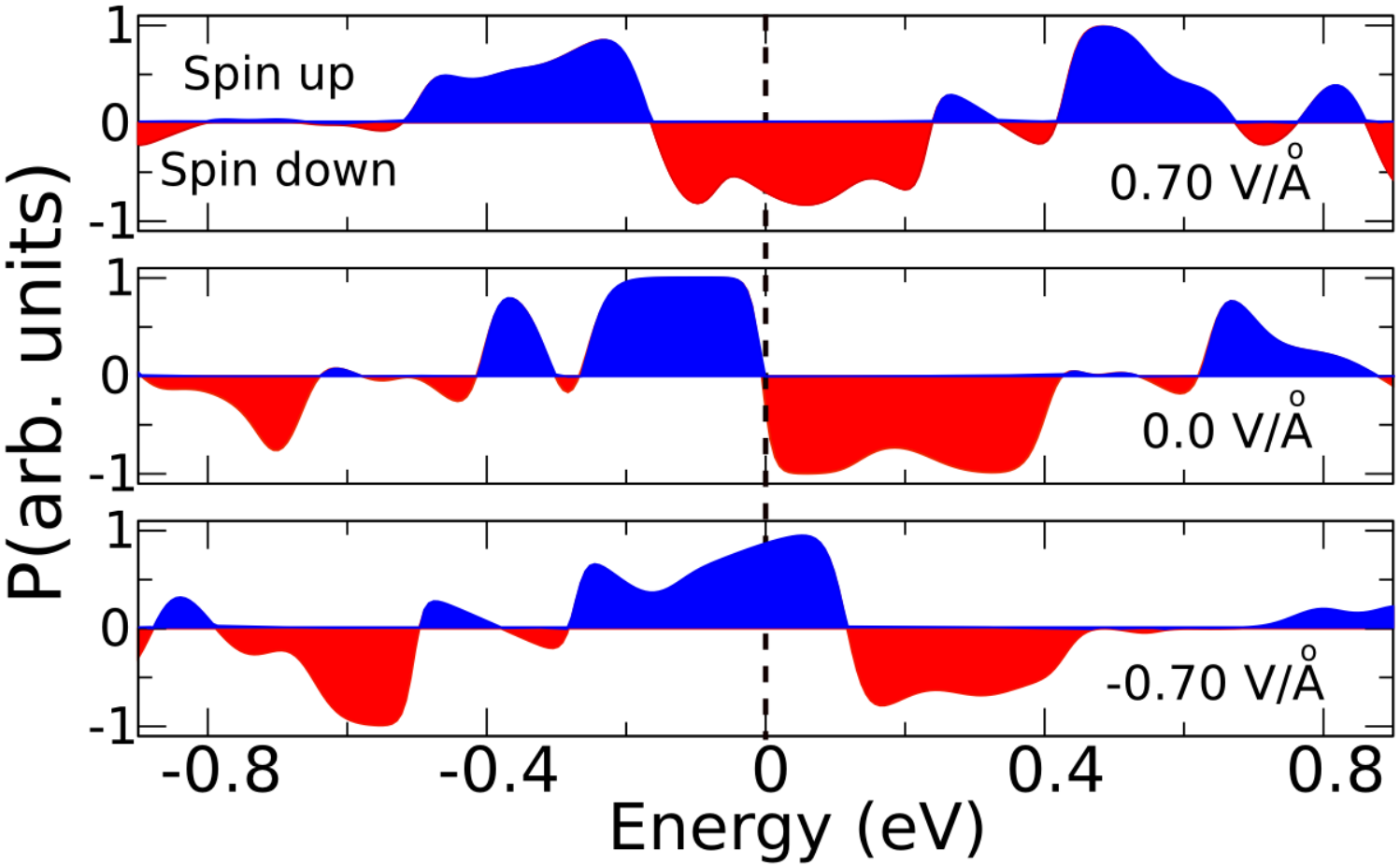
Magnetoelectric coefficient: $\alpha = \mu_0 \frac{\Delta S}{\Delta E}$

Same order of magnitude as in ferromagnetic films: Fe (001), Ni(001) and Co(001)

Interplay of defect-level and electric field on the electronic structure



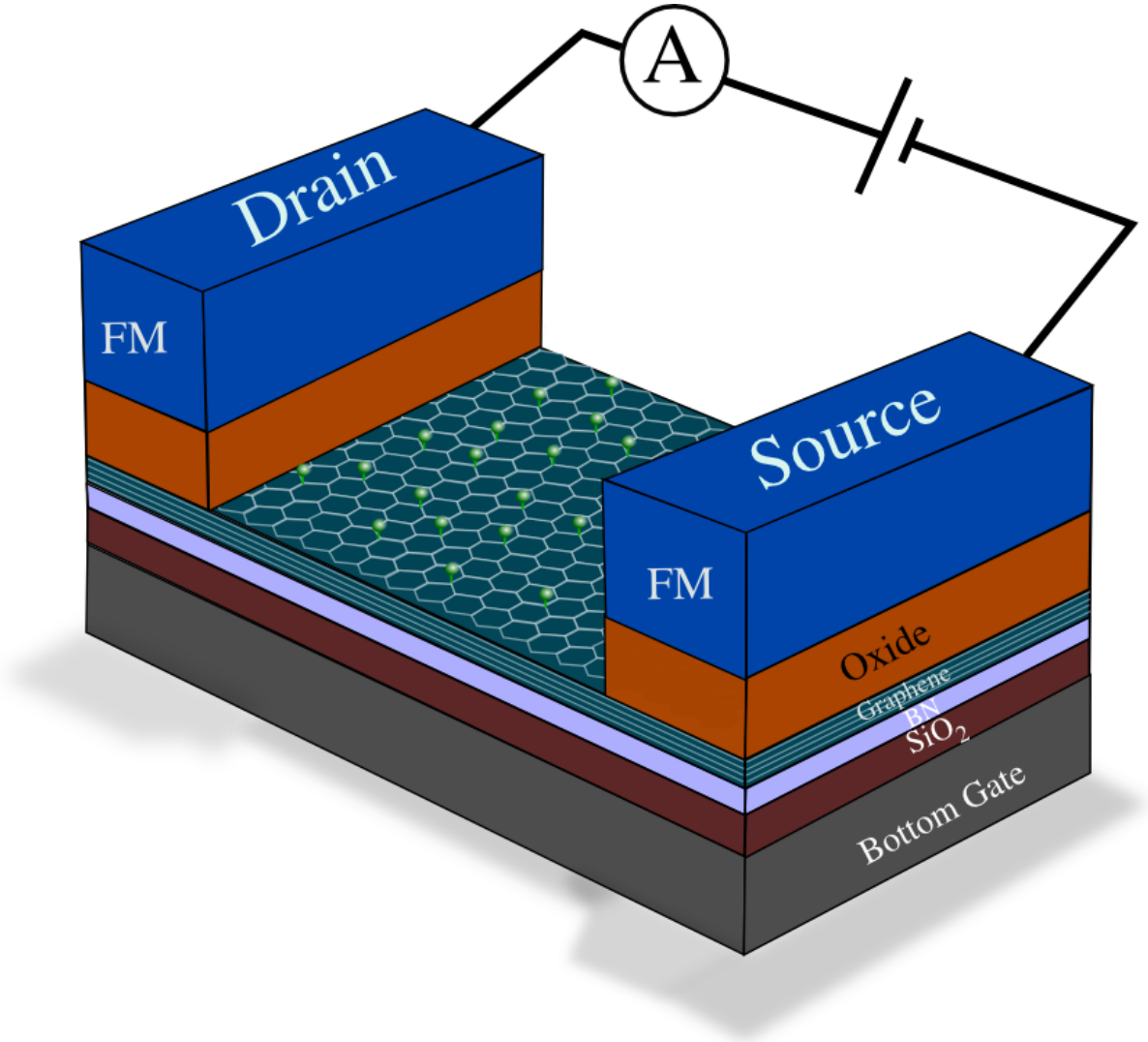
Doping-induced half-metallic behavior in graphene bilayer



$$P = \frac{N_{\uparrow} - N_{\downarrow}}{N_{\uparrow} + N_{\downarrow}}$$

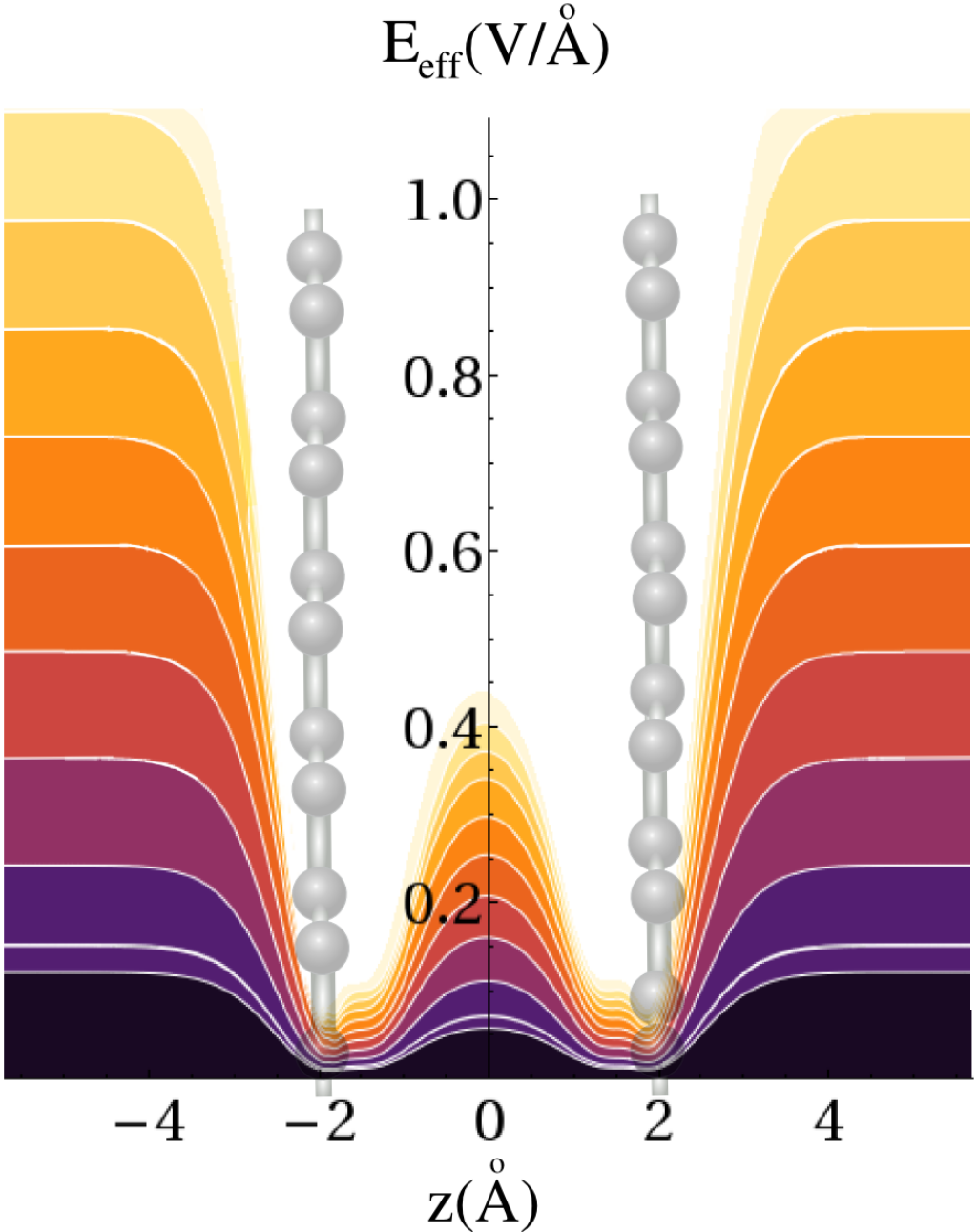
Selection of the spin-channel with an electric bias

Proposal of an experimental setup to check our predictions



Dielectric properties of bilayer graphene

See talk by E. J.G. Santos
on Wed. March 20
R6.00008

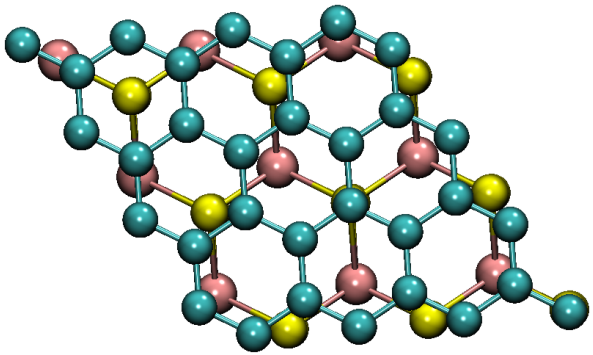


$$\varepsilon_G = \mathbf{E}_{ext}/\mathbf{E}_{eff}$$

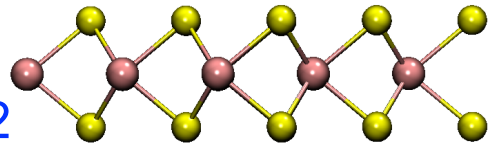
Position-dependent
interlayer electric field

Implications on e-e
interactions
on bilayer graphene

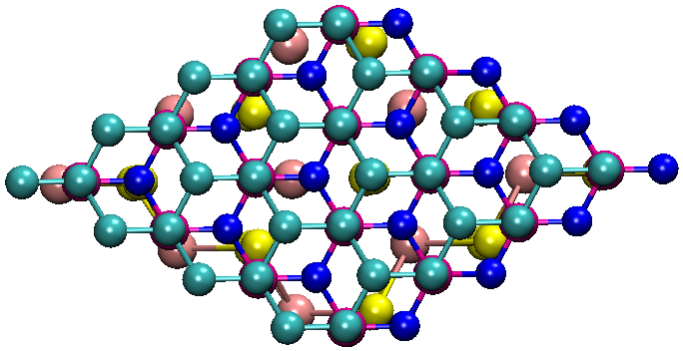
Work in progress:



G



MoS_2



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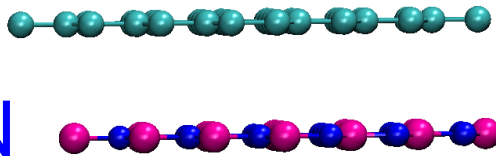
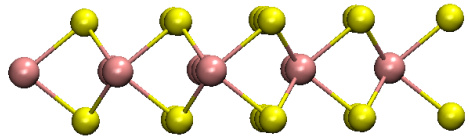
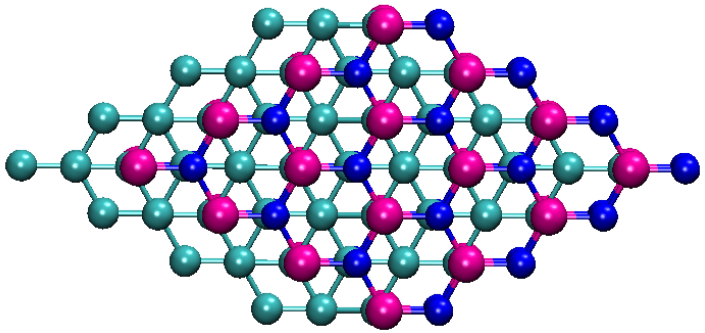


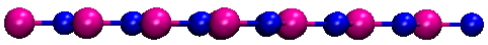
h-BN

MoS_2





h-BN



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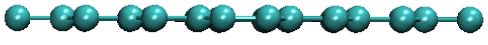

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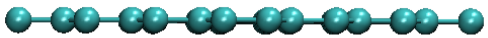
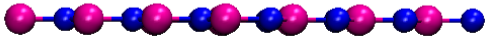
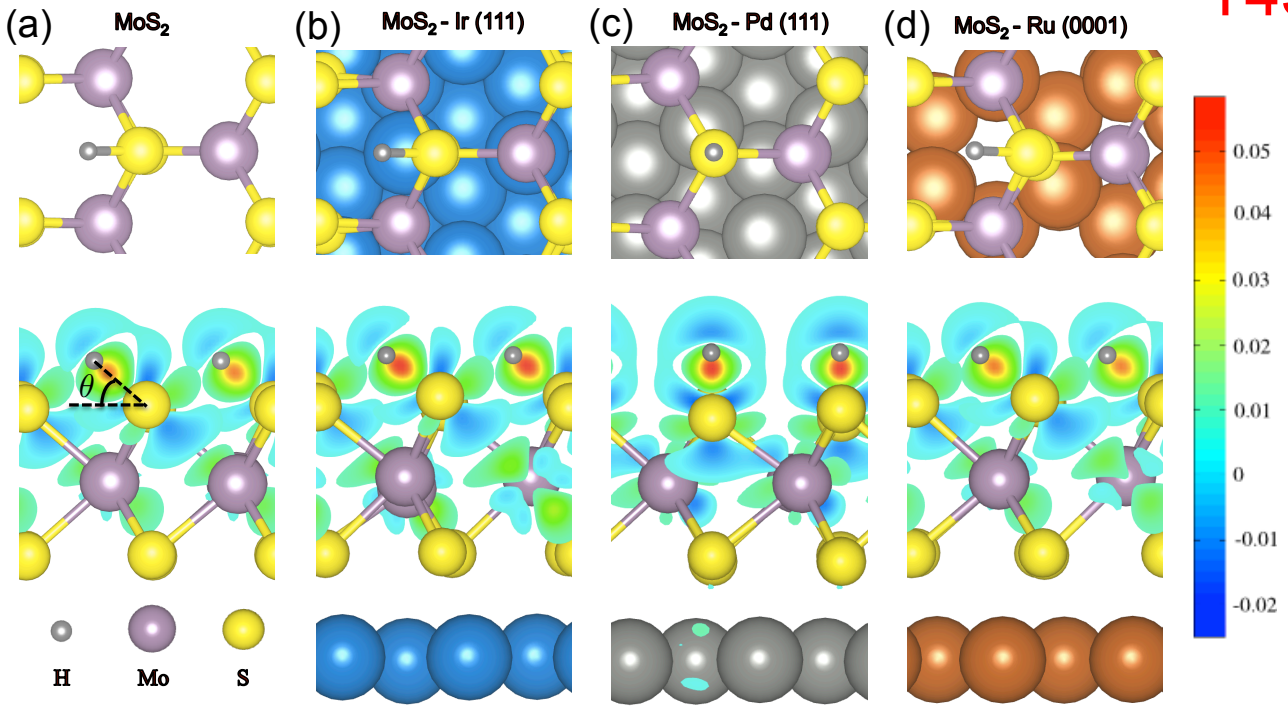


h-BN



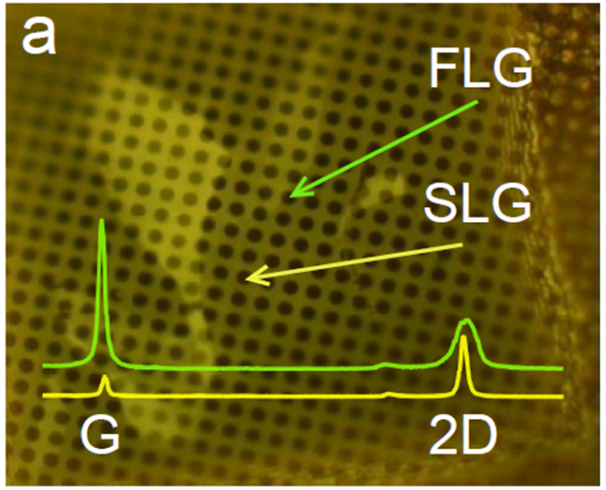
Hydrogen Adsorption on MoS_2/on metal surfaces

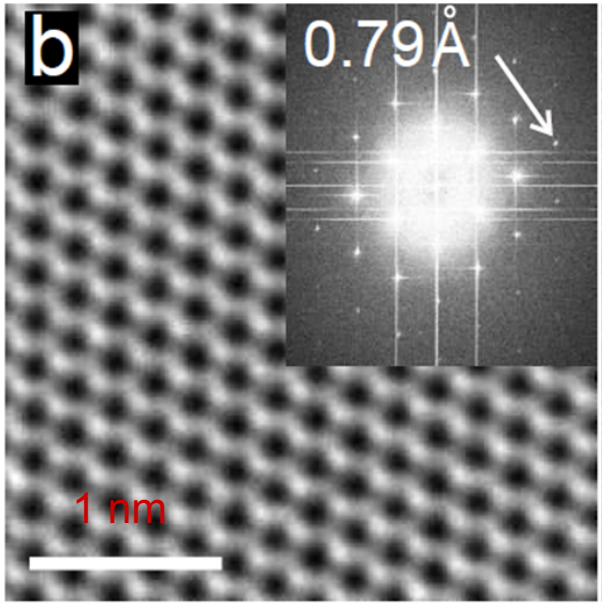
See talk by Wei Chen on Thu. March 21 T43.00012

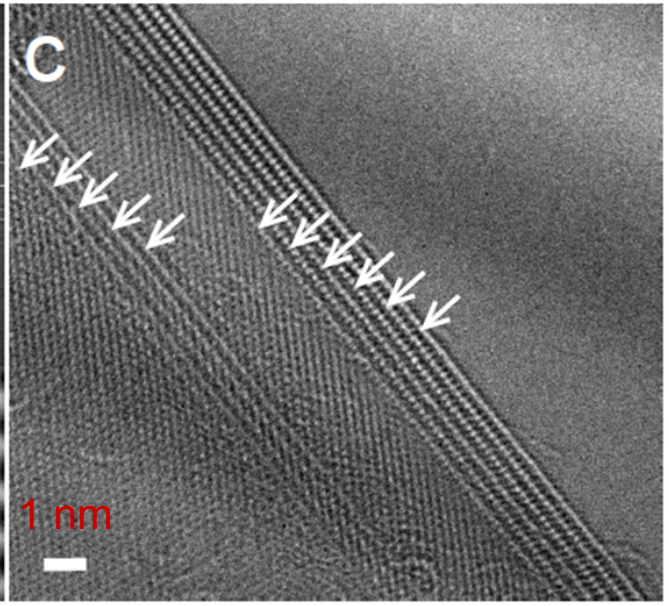


	E_b (eV)	d_z^0 (Å)	d_z^H (Å)	E_a (eV)	L_{H-S} (Å)	θ (deg)
free-standing MoS_2				1.07	1.46	40.2
MoS_2/Ir(111)	0.62	2.23	2.20	1.44	1.43	37.2
MoS_2/Pd(111)	0.74	2.17	2.09	1.39	1.39	89.1
MoS_2/Ru(0001)	0.82	2.25	2.20	1.33	1.46	38.2

Real-space imaging of graphene (Wei Li Wang)

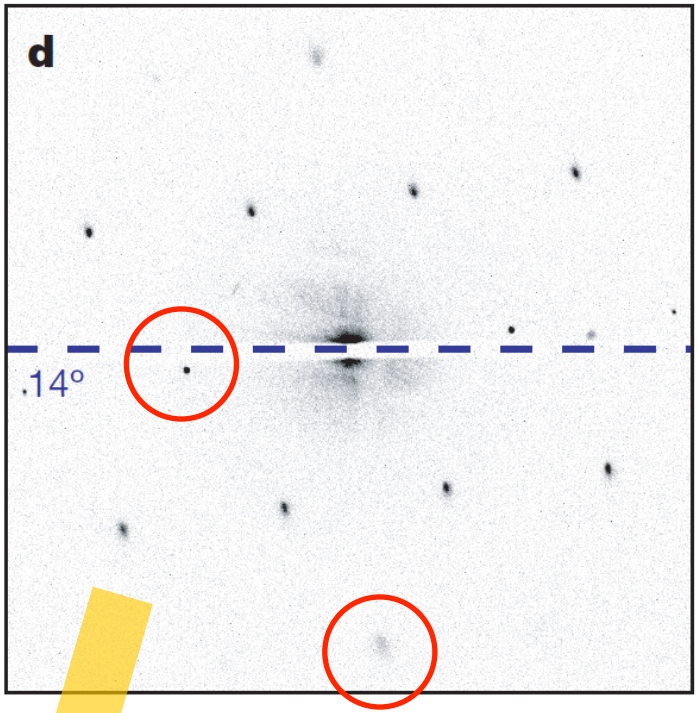


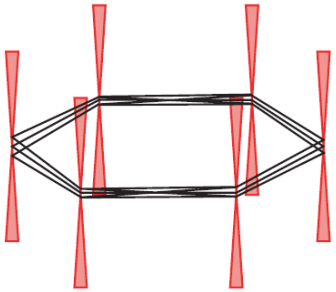


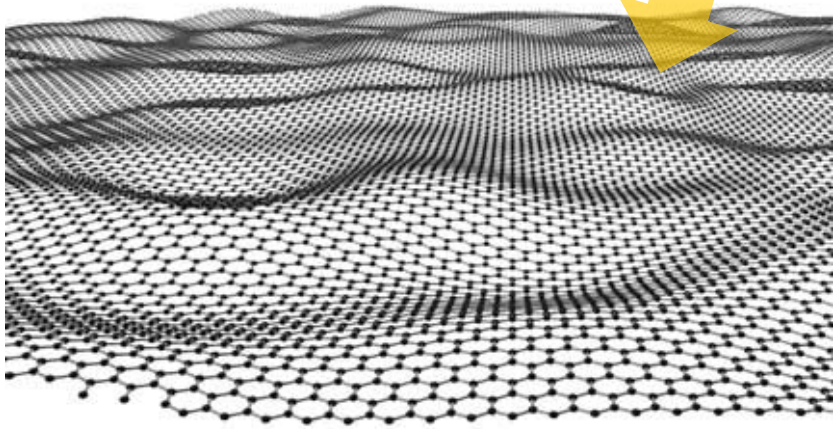


Libra MC 200-80
- Monochromated
- Aberration Corrected
- Operated at 80 kV

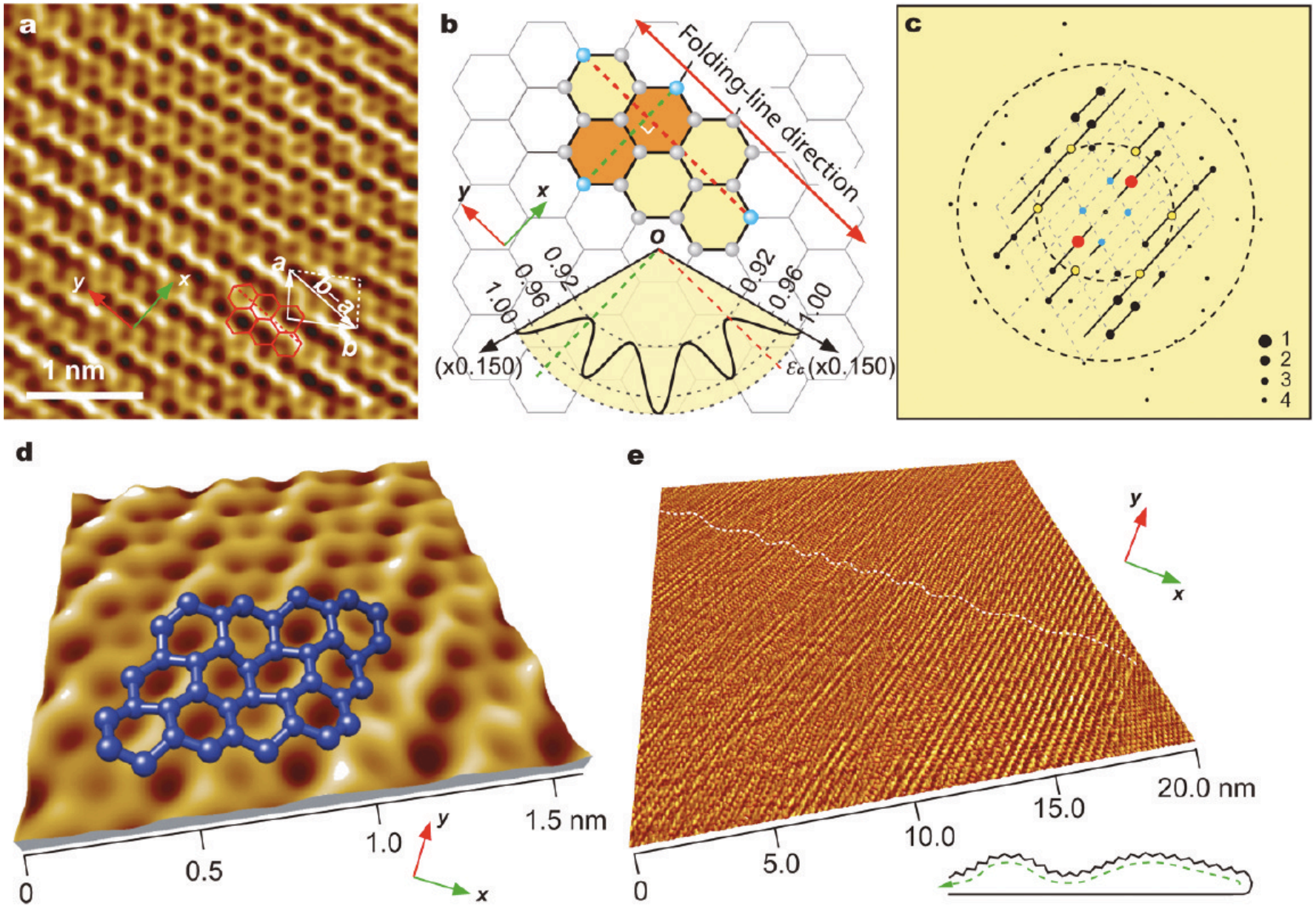
Intrinsic ripples in graphene







Experiment:
Meyer, J. C. et al. Nature **446**, 60-63, 2007.
Theory:
Mermin, N. D. Phys. Rev. **176**, 250–254, 1968.
Nelson, D. R., Piran, T. & Weinberg, *World Scientific*, Singapore, 2004.

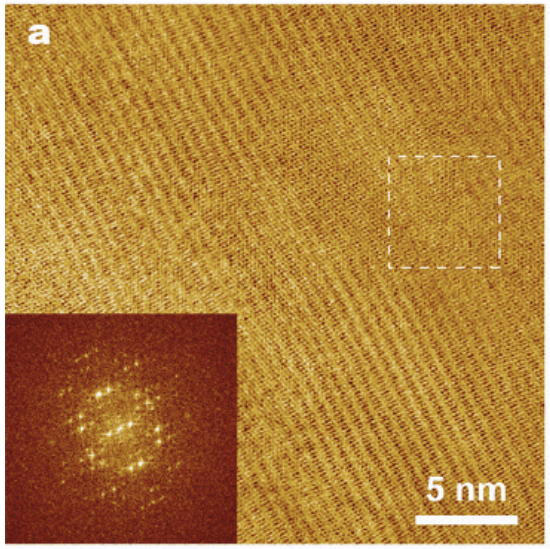


Graphene Structures at an Extreme Degree of Buckling

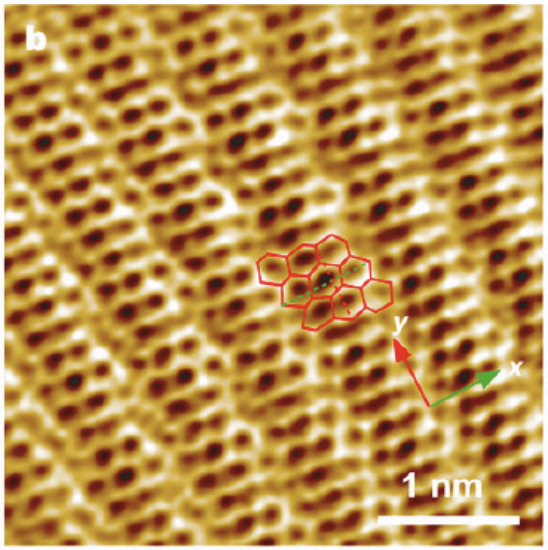
Youdong Mao, Wei L. Wang, Dongguang Wei, Efthimios Kaxiras, and Joseph G. Sodroski

ACS NANO, **5**, 1395–1400 (2011)

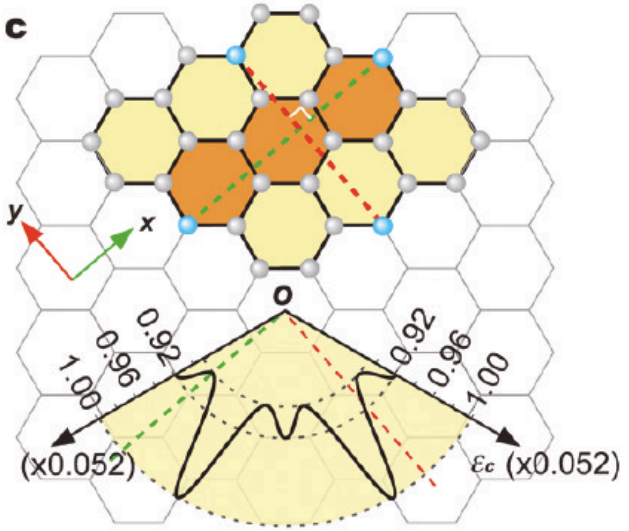
TEM

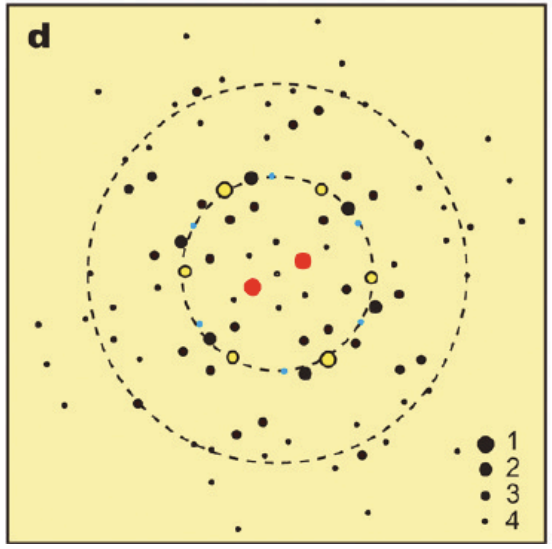


TEM

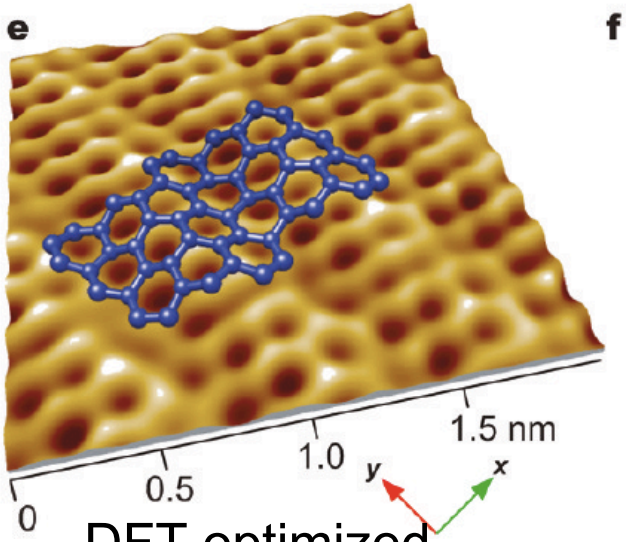


Atomic model

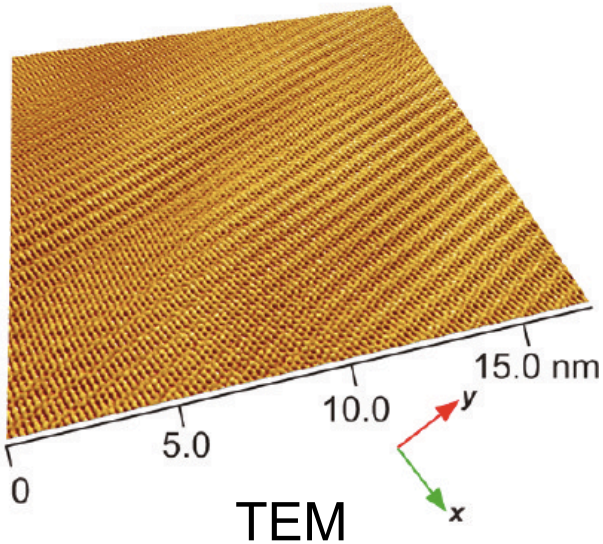




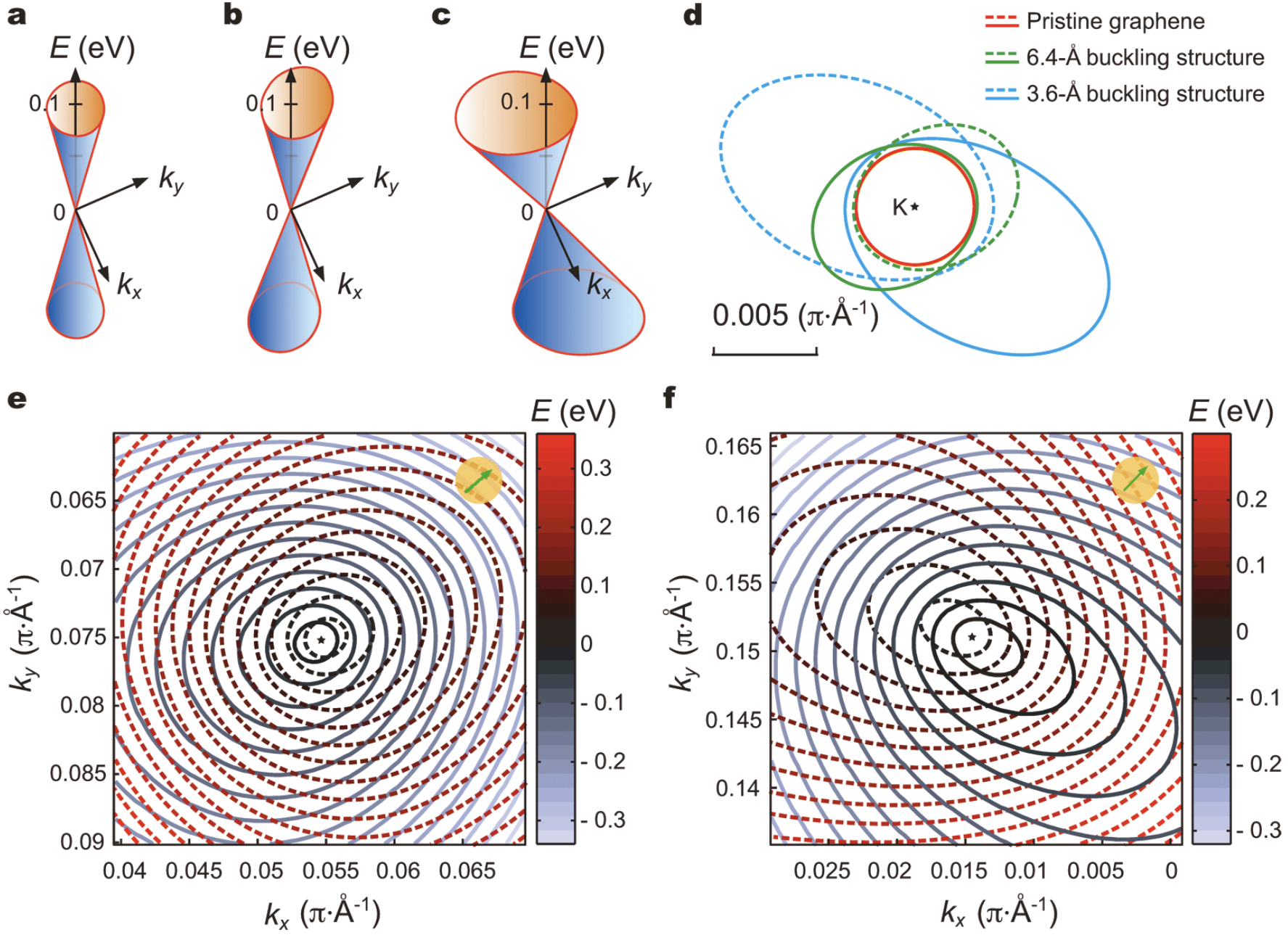
Fourier spectrum



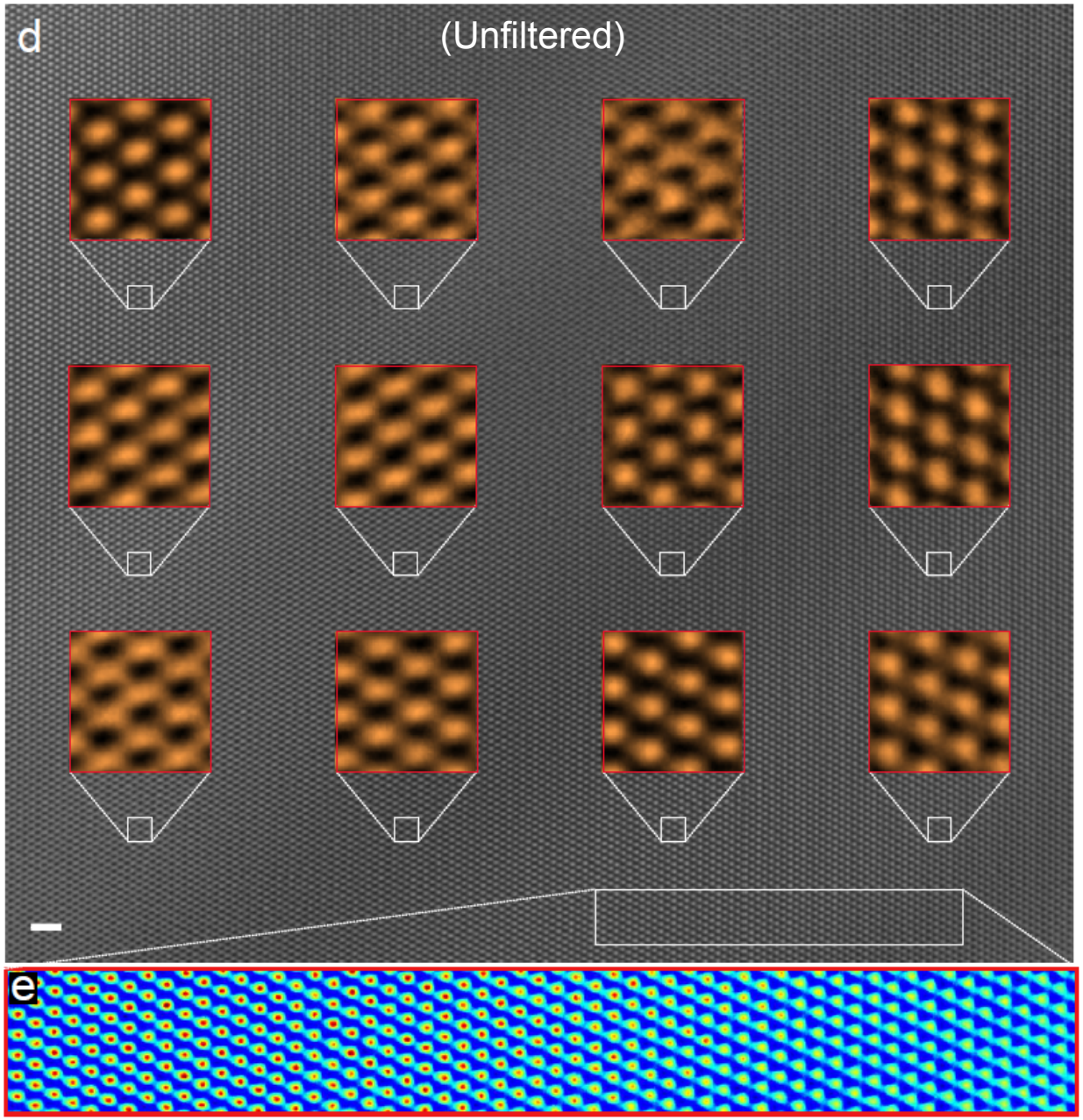
DFT-optimized structure



TEM


a
b
c
d
E (eV)
0.1
0
k_y
k_x
Pristine graphene
6.4-Å buckling structure
3.6-Å buckling structure
K
0.005 (π·Å^{-1})
e
f
E (eV)
0.3
0.2
0.1
0
- 0.1
- 0.2
- 0.3
k_y (π·Å^{-1})
k_x (π·Å^{-1})
0.065
0.07
0.075
0.08
0.085
0.09
0.04
0.045
0.05
0.055
0.06
0.065
0.165
0.16
0.155
0.15
0.145
0.14
0.025
0.02
0.015
0.01
0.005
0

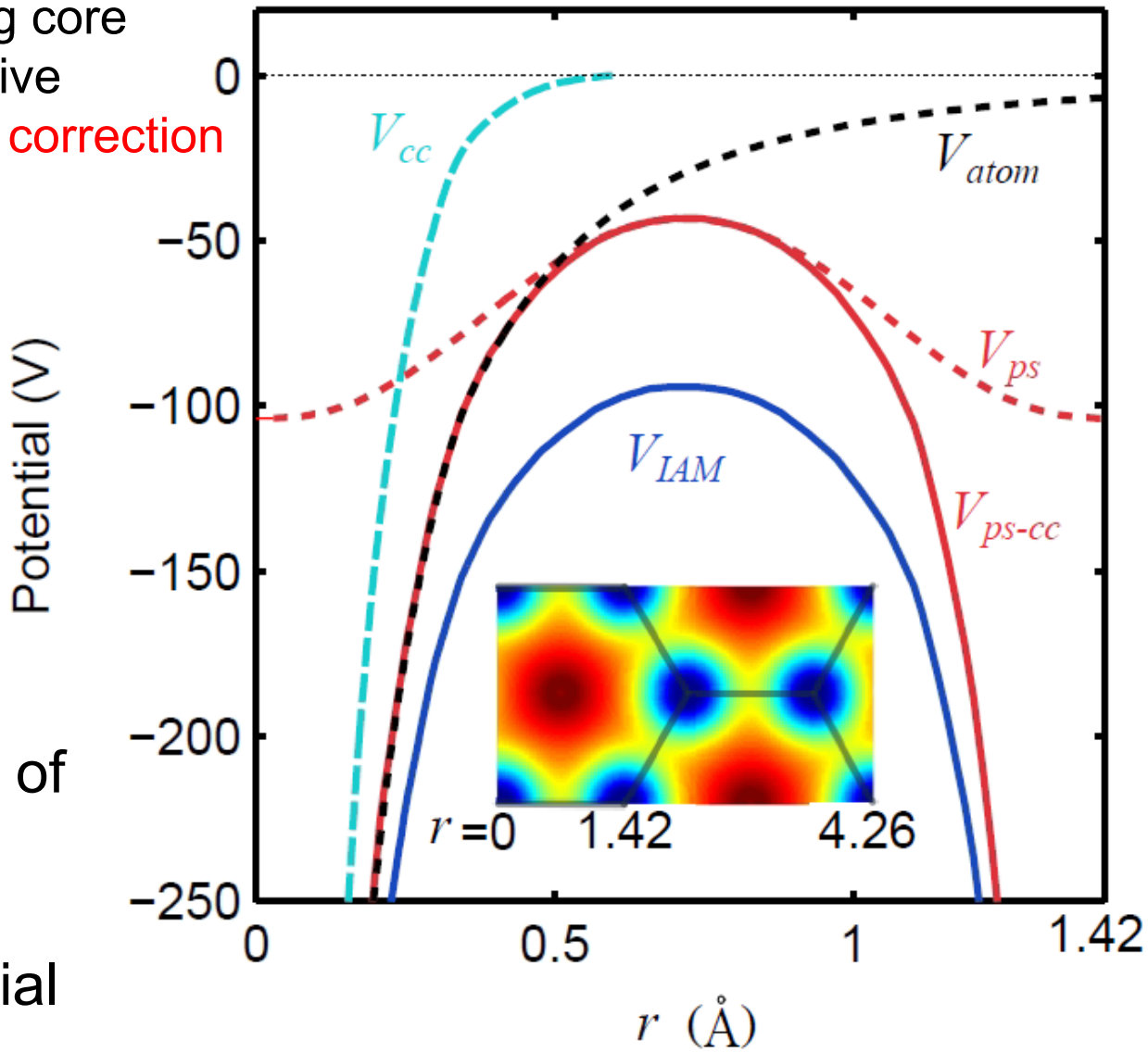
Ripples in real space



Calculate total potential with a transferable core correction

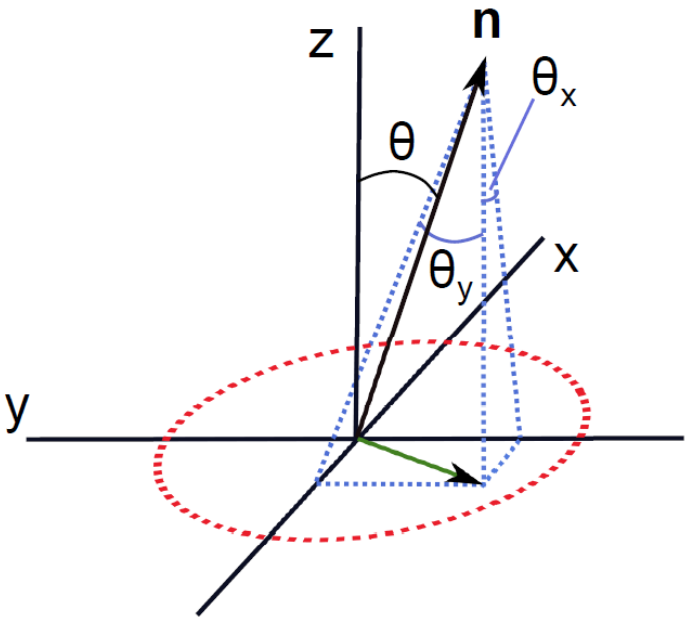
DFT:

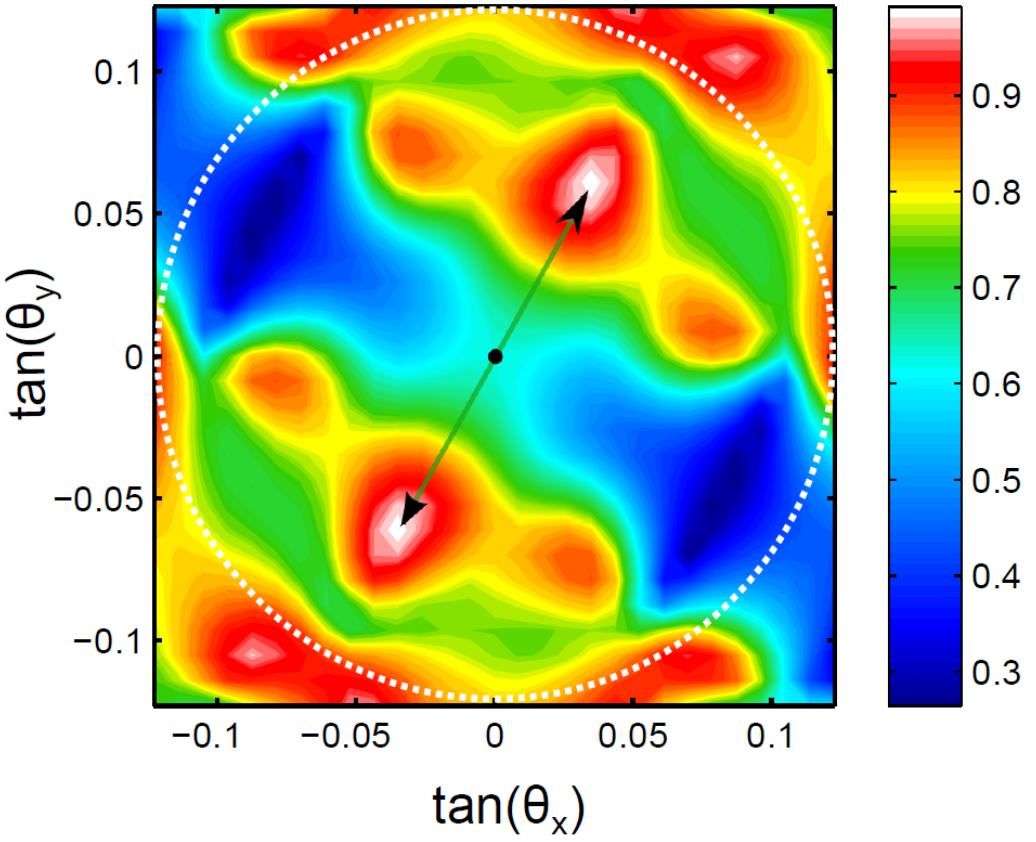
- Pseudopotential : wrong core
- All electron: too expensive
- Pseudopotential + core correction



Simulate scattering of incident electron beam by total electrostatic potential

Ambiguity: inversion symmetry



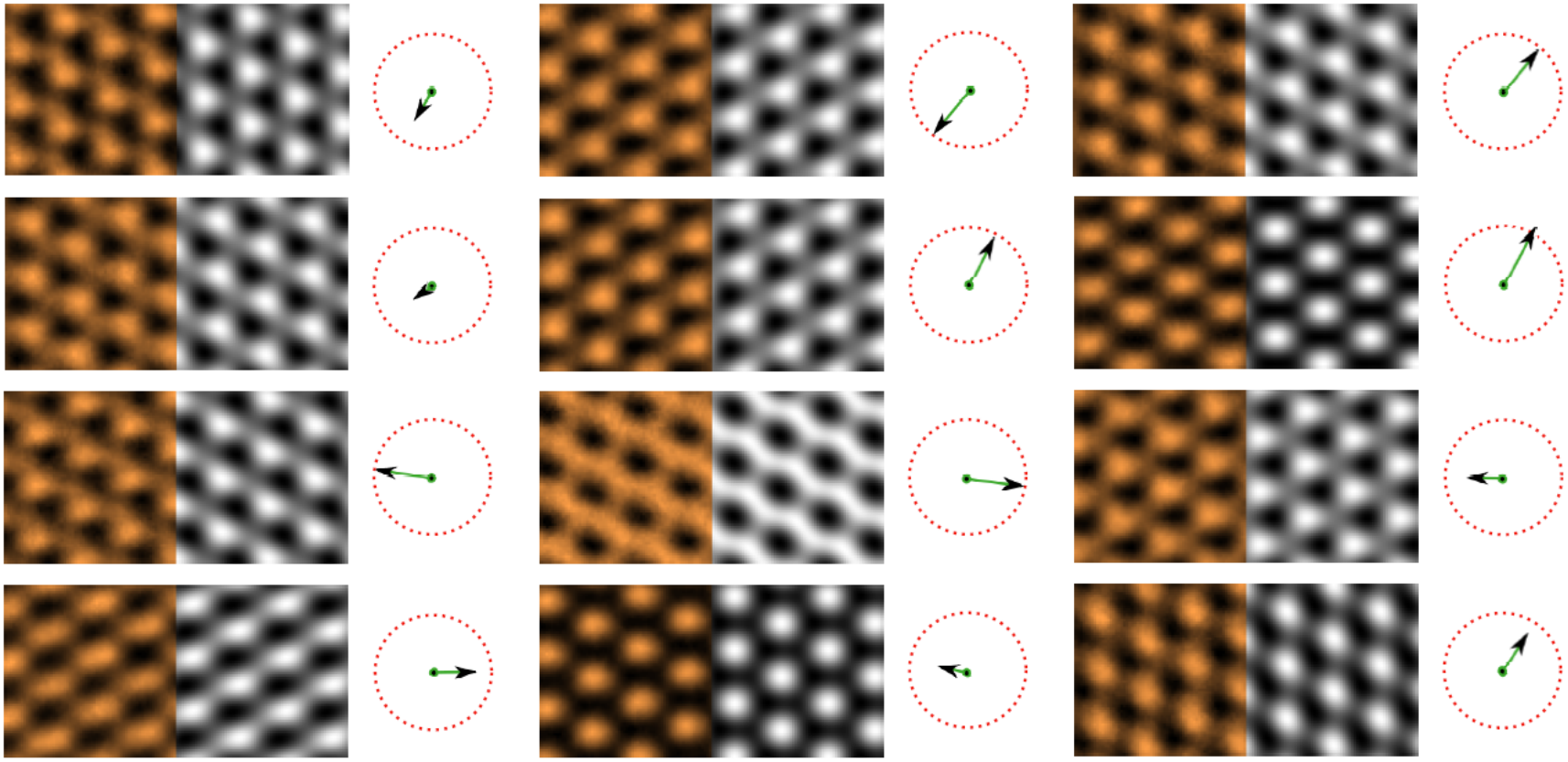


Matching based on a normalized cross-correlation functional:

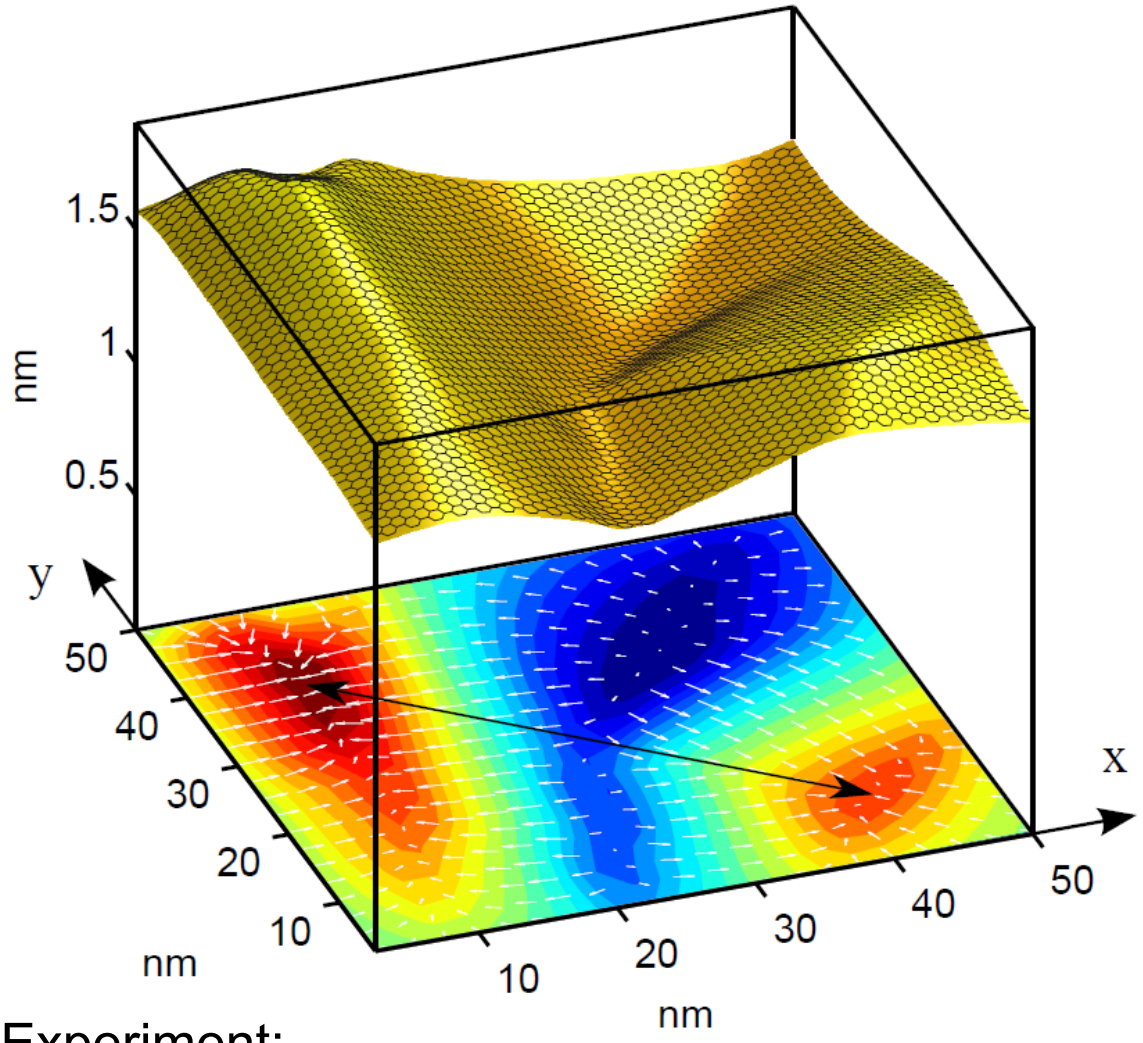
$$C(f_{exp}, f_{sim}) = \frac{\sum_{xy}(f_{exp}(x,y) - \overline{f}_{exp})(f_{sim}(x,y) - \overline{f}_{sim})}{\sqrt{\sum_{xy}(f_{exp}(x,y) - \overline{f}_{exp})^2 \sum_{xy}(f_{sim}(x,y) - \overline{f}_{sim})^2}}$$

Kirkland, E. J. *Advanced computing in electron microscopy* (Springer, 2010).

Image matching



Topography Reconstruction



Amplitude: 0.5 nm
Std. dev.: 0.13 nm
Typical width: 45 nm

NanoLetters (2012)

Experiment:

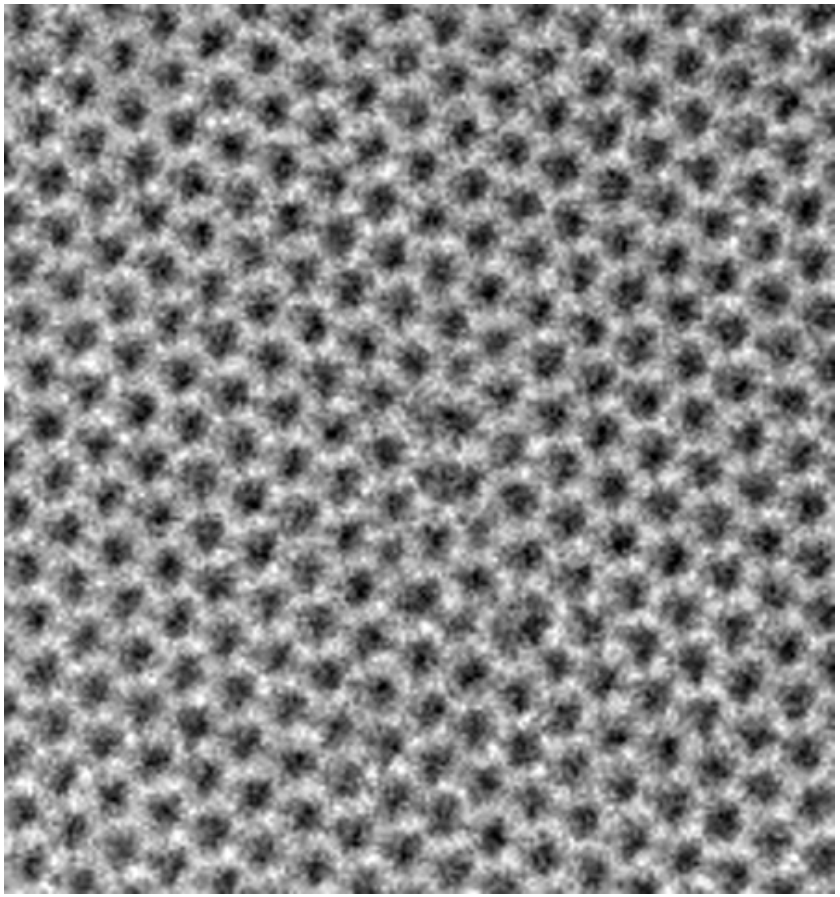
- Wei Li Wang, D. Bell, Wei Yi, S. Bhandari, R. Westervelt

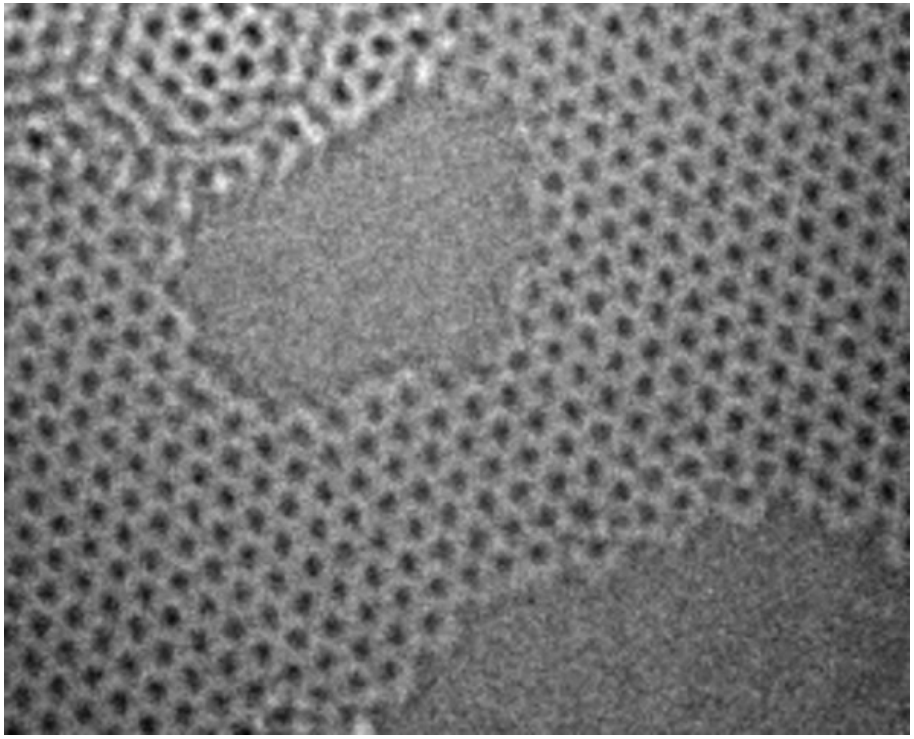
Theory:

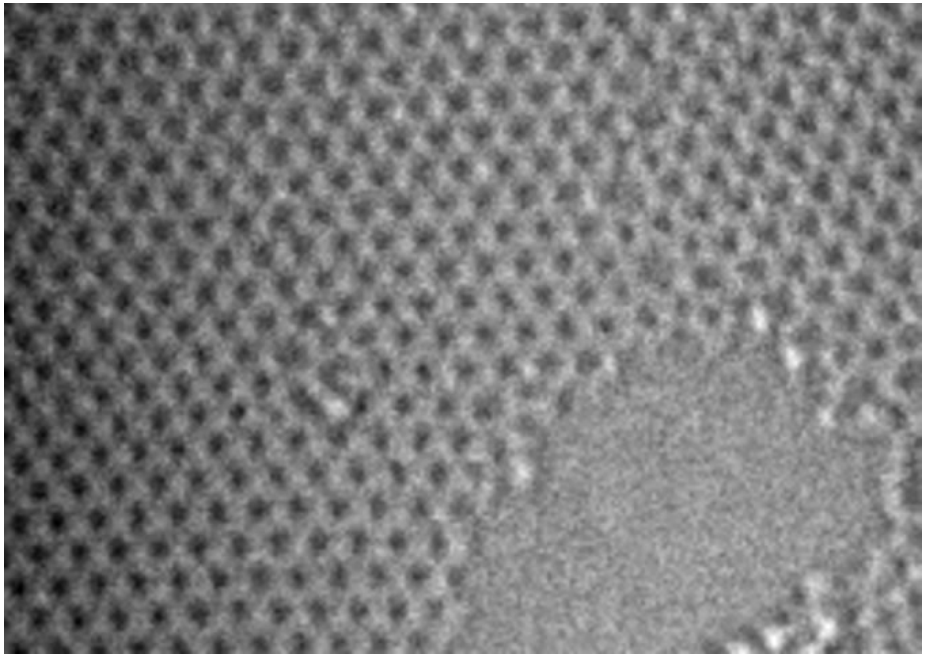
- Wei Li Wang, E. G.J. Santos, E.Kaxiras

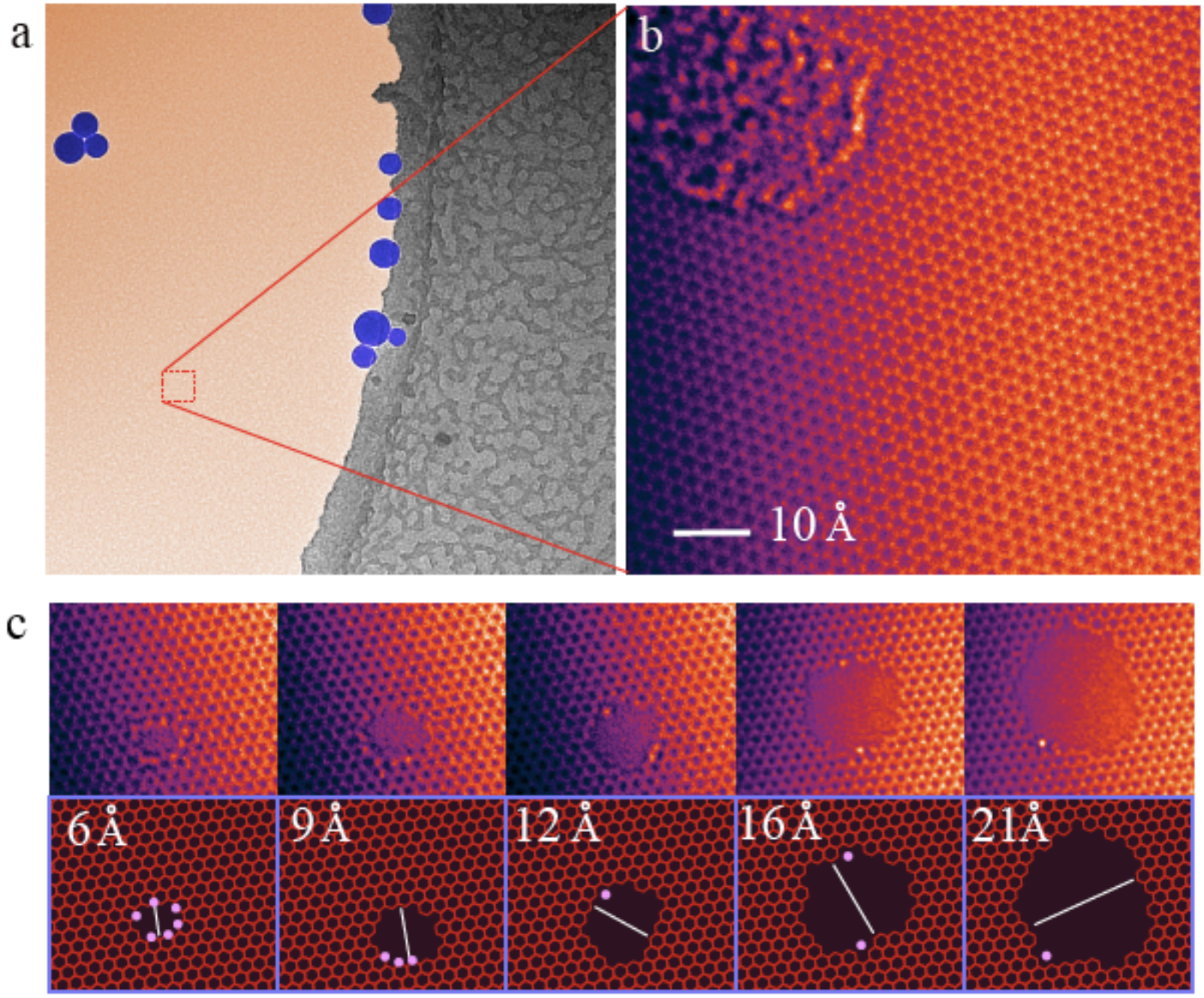
An atomic-scale chisel for sculpting graphene

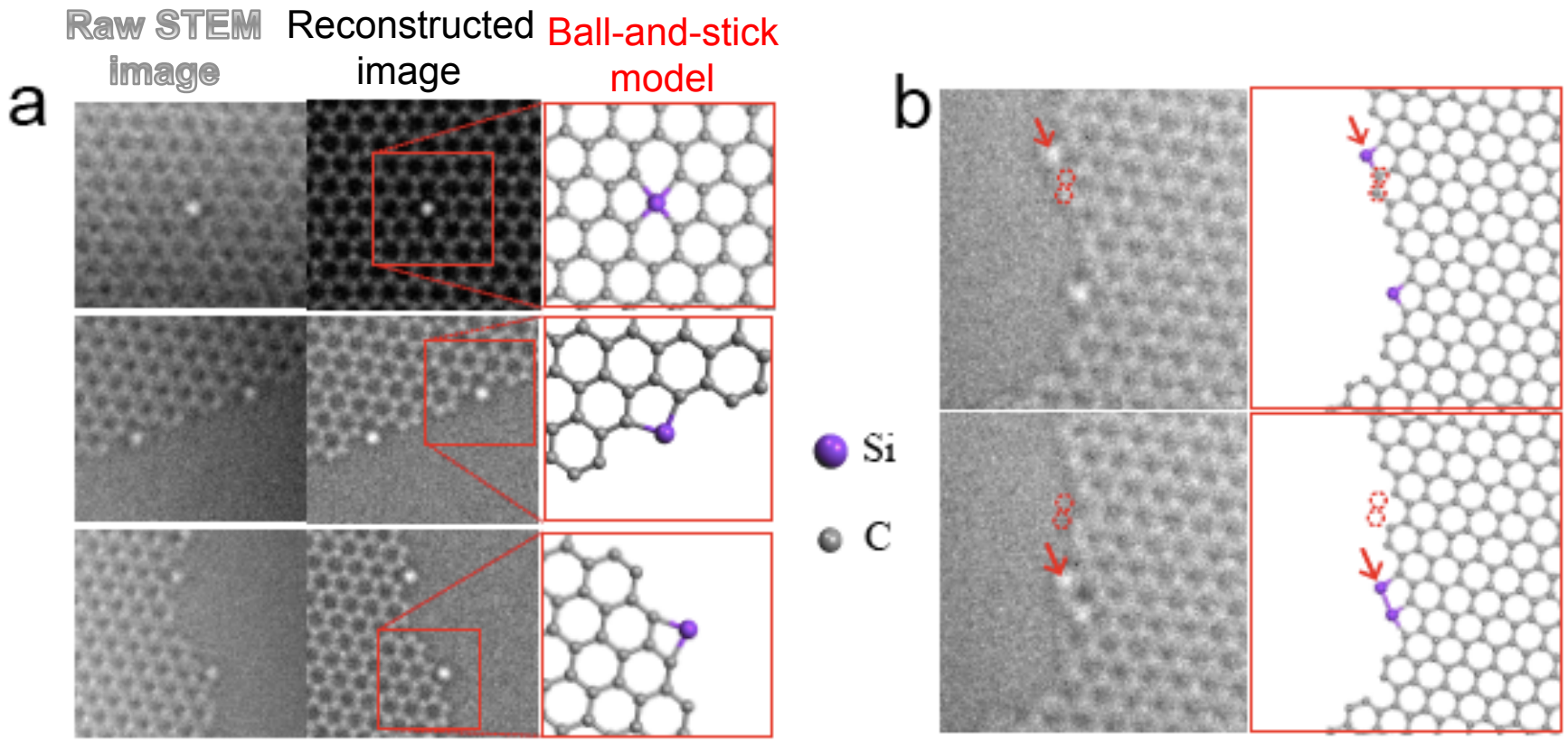
By Wei Li Wang
Expts. carried out at LBNL's NCEM

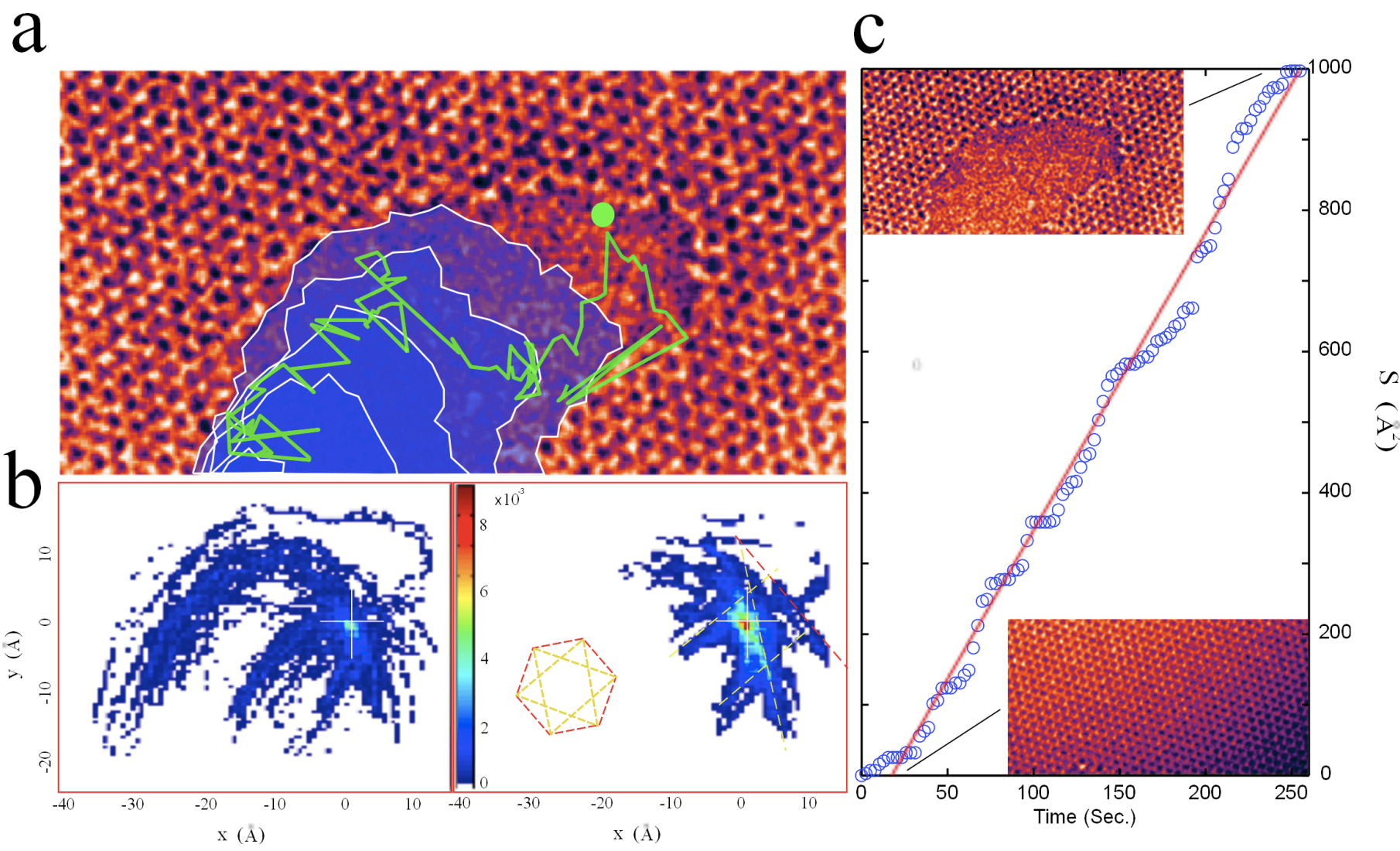






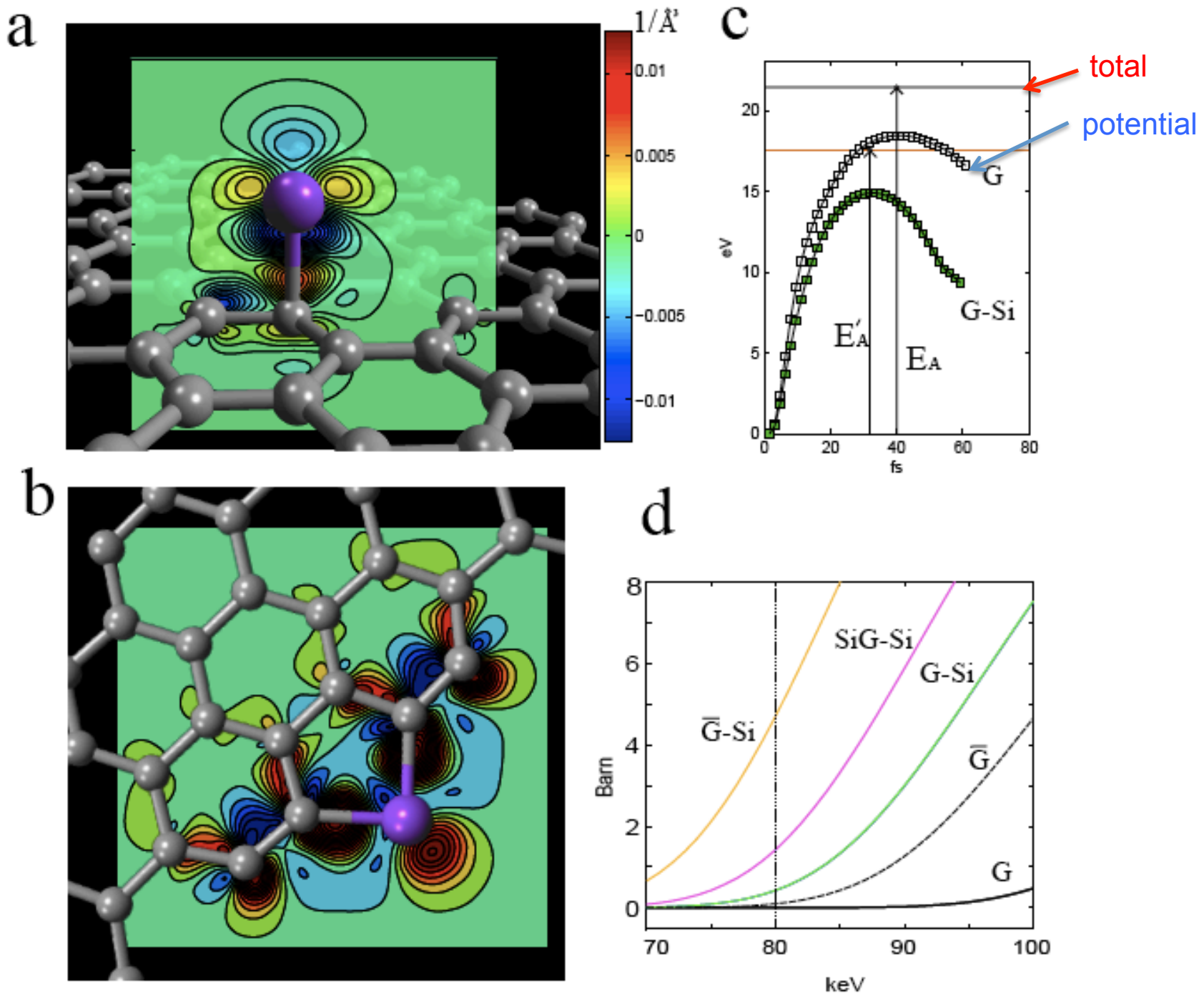

a
b
10 Å
c
6 Å
9 Å
12 Å
16 Å
21 Å


a
Raw STEM image
Reconstructed image
Ball-and-stick model
Si
C
b



$\mathbf{a} = \mathbf{r}_p - \mathbf{r}_{Si}$

$\mathbf{b} = \mathbf{r}_m - \mathbf{r}_{Si}$


a
1/Å³
0.01
0.005
0
−0.005
−0.01
b
c
total
potential
G
G-Si
E′A
EA
eV
fs
0
5
10
15
20
0
20
40
60
80
d
SiG-Si
G-Si
G
G-Si
G
Barn
keV
0
2
4
6
8
70
80
90
100