Tailoring Electronic Coupling in Single-Crystal Bilayer Graphene

NCKU (July 5, 2012)

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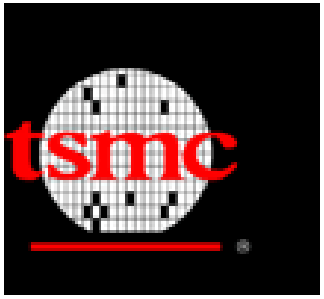


National Tsing Hua University

Particularly thanks the hard-working group members…

Also Dr. Kazu Suenaga…

Outline

1. About graphene growth…
   - CVD graphene
   - CVD graphene without grain boundaries

2. About bilayer graphene …
   - Tailoring a twist in bilayer
   - TEM + Raman technique
   - Coupling or decoupling?

First graphene growth on Ni

1970-1980. Jack Blakely and team: Graphene Growth on Metals*

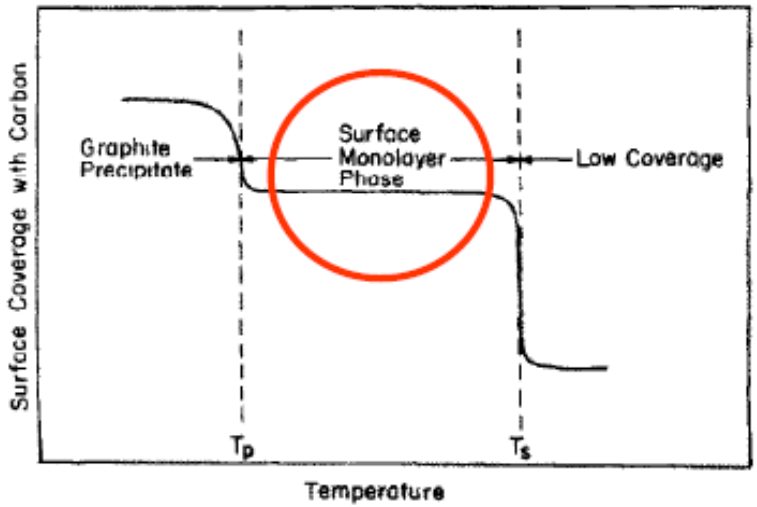
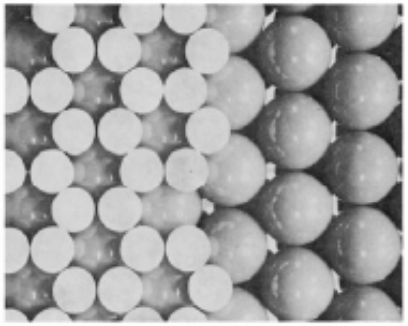


Fig. 1. Schematic equilibrium temperature dependence of carbon coverage on the (111) surface of a carbon doped nickel single crystal. A phase transition from a low coverage to a condensed state occurs at the segregation point T_s; graphite precipitation starts at T_p.



Ball model of a graphite(0001) layer on the (111) surface of Ni. The left hand portion of the photographs depicts the probable arrangement in state B with (0001)graphite ‖(111)Ni; [11$\bar{2}$0]graphite ‖[1$\bar{1}$0]Ni. Note that the graphite layer has two atoms per unit mesh.

Graphitic carbon monolayer formation by surface segregation on Ni (111),(110),(311); Pd (100),(111); Co (0001).

Shelton JC, Patil HR, Blakely JM. *Surf. Sci.* **43**:493 (1974); Eizenberg M and Blakely JM. *J. Chem. Phys.* **71**: 3467 (1979); Eizenberg M and Blakely JM. *Surf.Sci.*, **82**:228 (1979); Hamilton JC and Blakely JM. *Surf. Sci.* **91**:119 (1980)

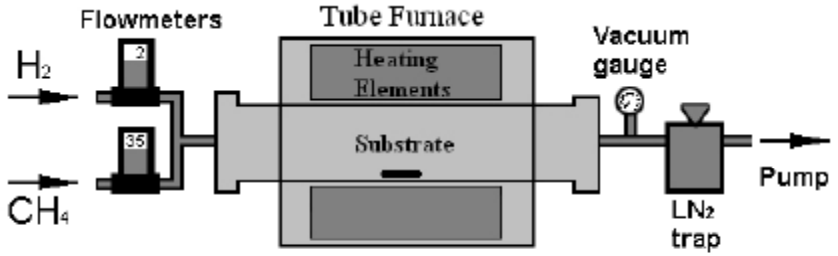
But: Isolation/transfer of graphene from metal substrates onto other substrates is quite recent. Yu *et al. Appl. Phys. Lett.* **93**:113103 (2008); Reina *et al.* Nano Lett. **9**:30 (2009); Li et al Science **324**: 1312-1314 (2009)

First graphene growth on Cu

Large-Area Synthesis of High-Quality and Uniform Graphene Films on Copper Foils

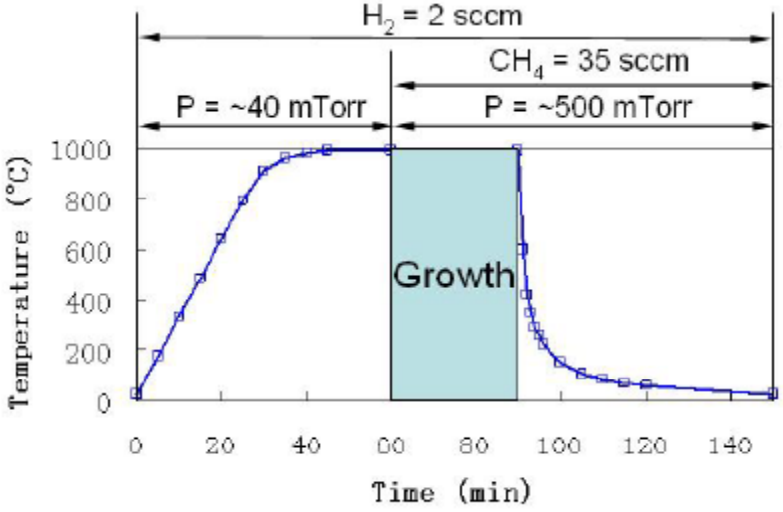
Xuesong Li,[1] Weiwei Cai,[1] Jinho An,[1] Seyoung Kim,[2] Junghyo Nah,[2] Dongxing Yang,[1] Richard Piner,[1] Aruna Velamakanni,[1] Inhwa Jung,[1] Emanuel Tutuc,[2] Sanjay K. Banerjee,[2] Luigi Colombo,[3]* Rodney S. Ruoff[1]*

5 JUNE 2009 VOL 324 SCIENCE



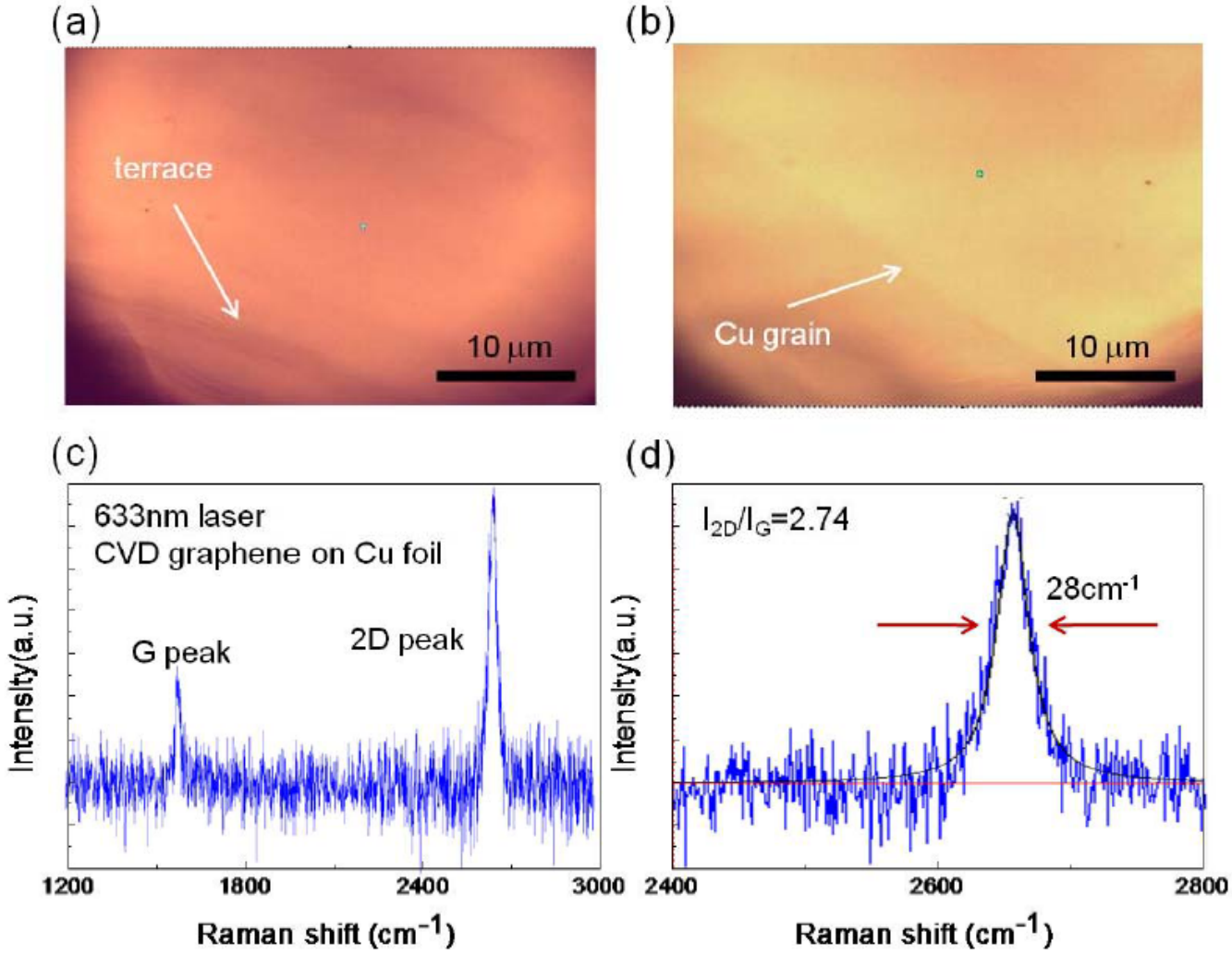
Graphene Growth Process

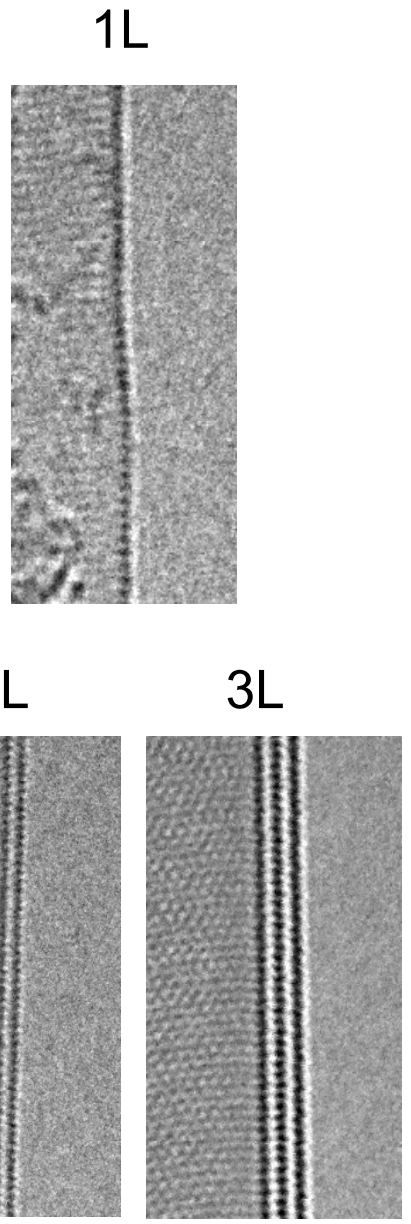
- 25 μm thick 99.8% pure Cu foil
- Load Cu foil in furnace
- Evacuate furnace
- Heat to T ~ 1040°C under H_2 gas
- Introduce CH_4 at a flow rate of 35 sccm and P = 500 mTorr
- Grow graphene for 1 to 20 min
- Cool to room temperature



Graphene grown on Cu

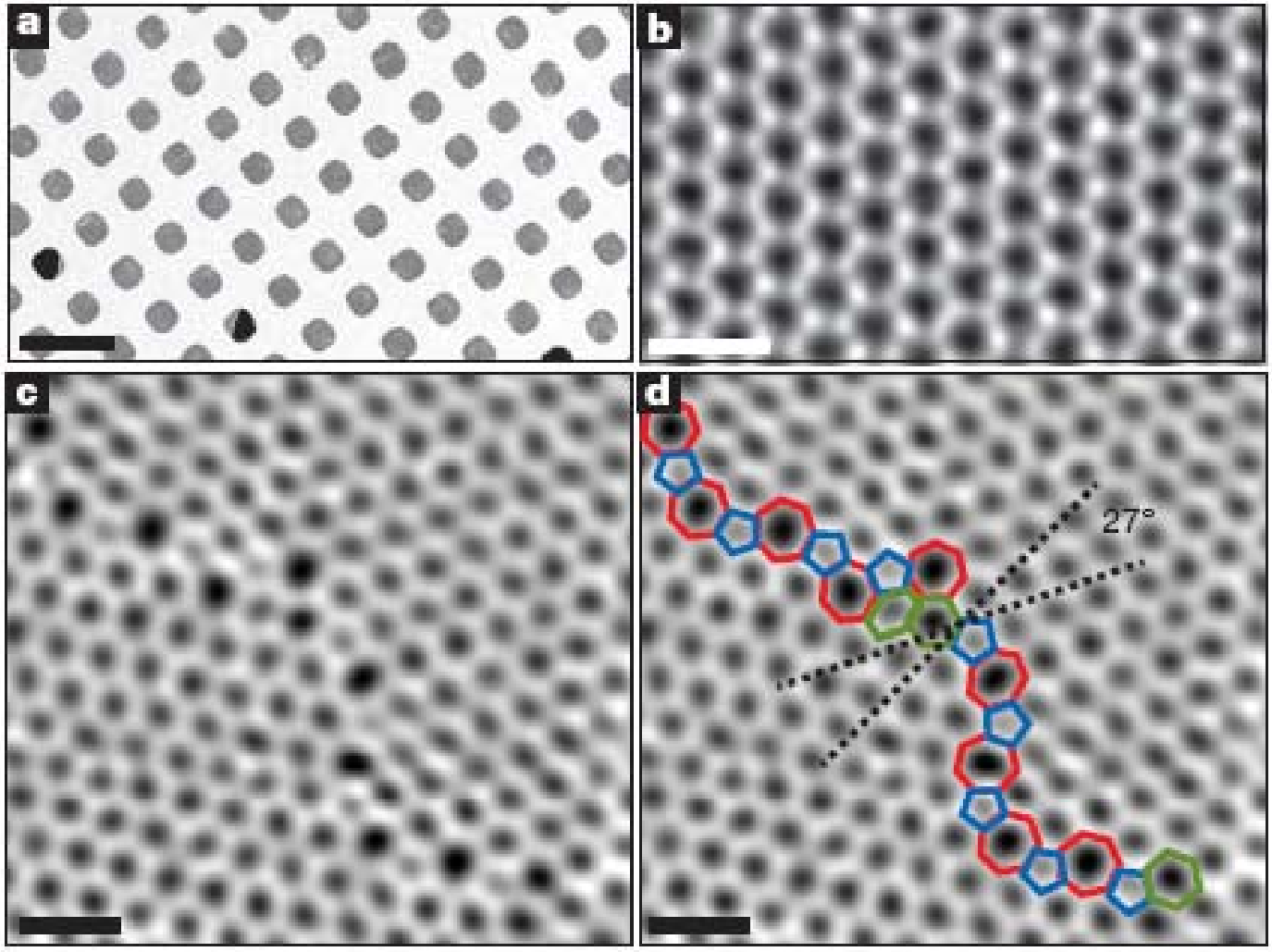
Over 90% of the films is one-layer graphene





C. C. Lu, *et al*., Langmuir, 27, 13748 (2011).

Polycrystalline structure



P. Y. Huang *et al.,* Nature 2011

Tuning the CVD on Cu

Control the layer number:

LPCVD

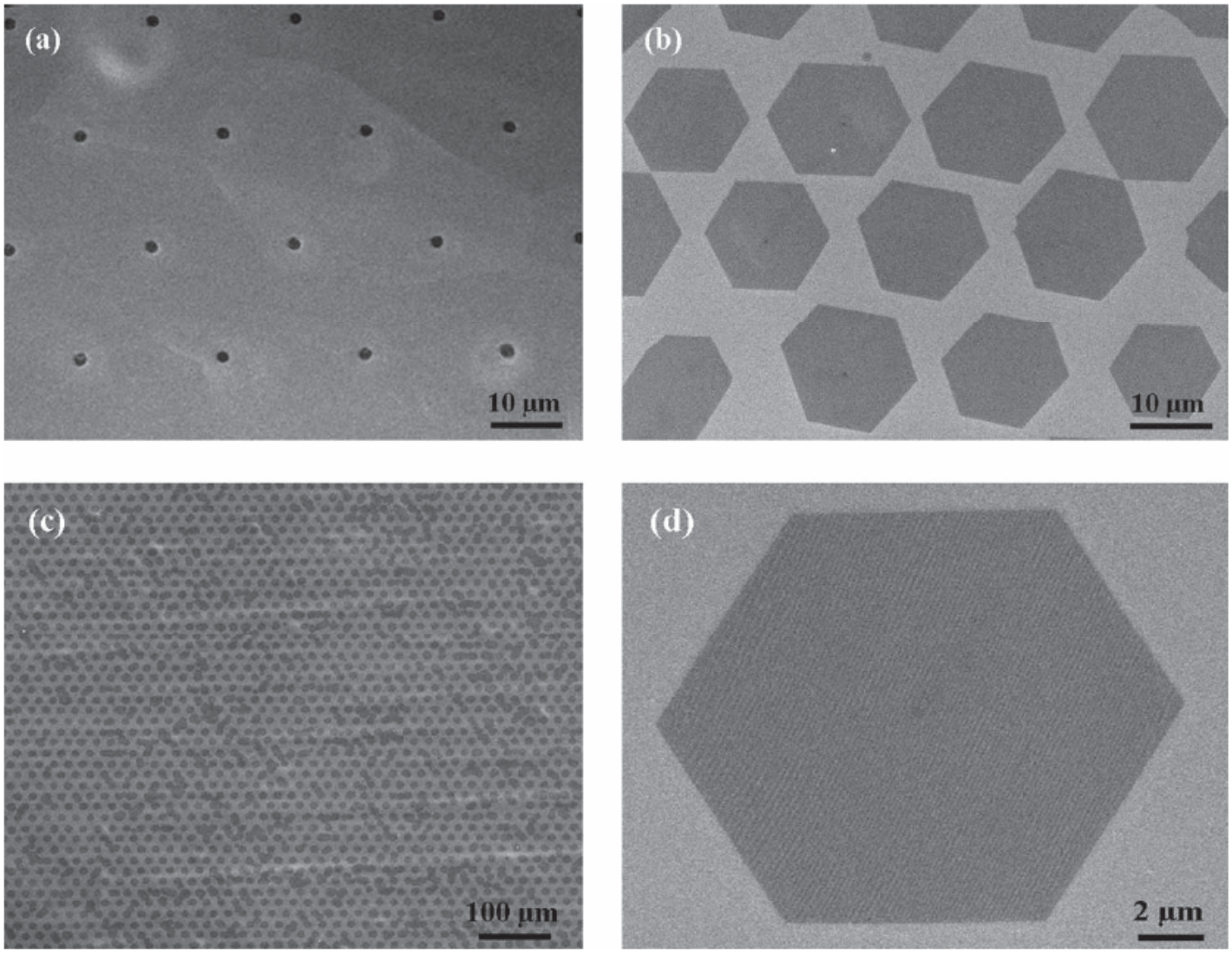
Self-limiting growth

Control the grain size:

APCVD with low C feedstock

High temperature

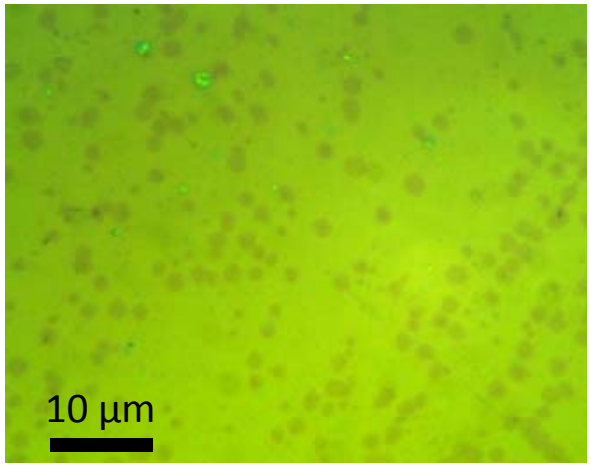
Single-crystal graphene on Cu (SEM)



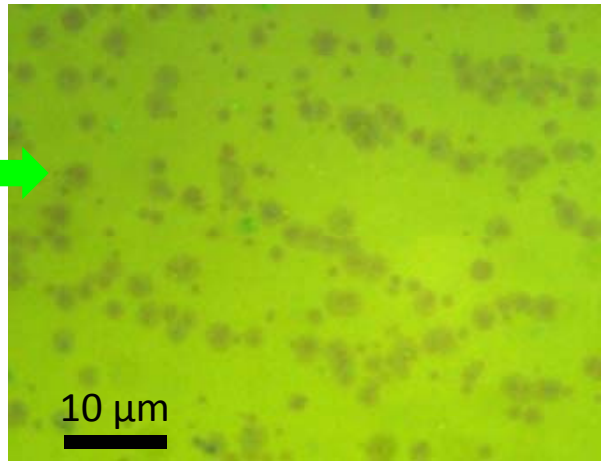
Adv. Mater. **2011**, *23*, 4898–4903

Single-crystal graphene on Cu (OM)

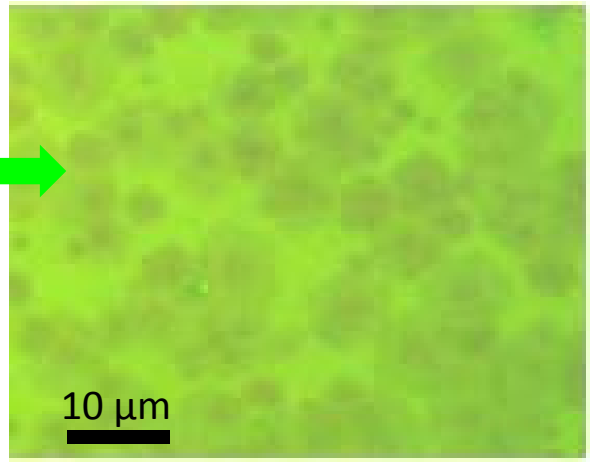
CH4: 1 SCCM
Growth: 3min40sec



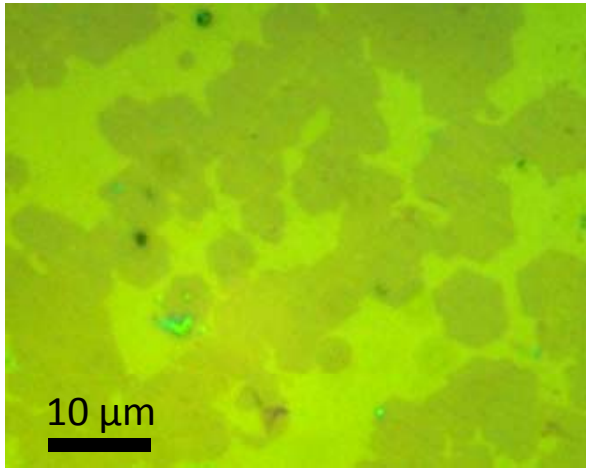
CH4: 1 SCCM
Growth: 4min



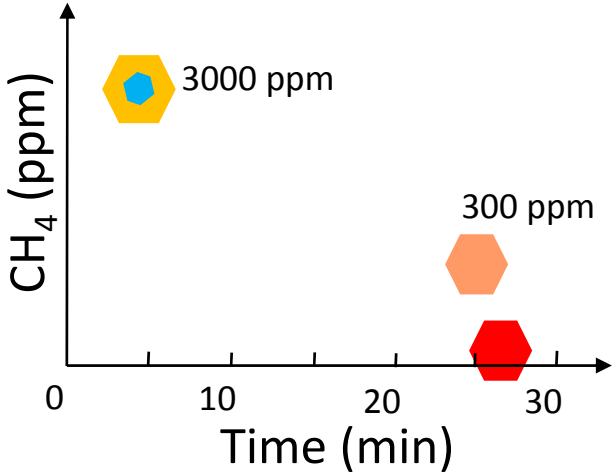
CH4: 1 SCCM
Growth: 4min30sec



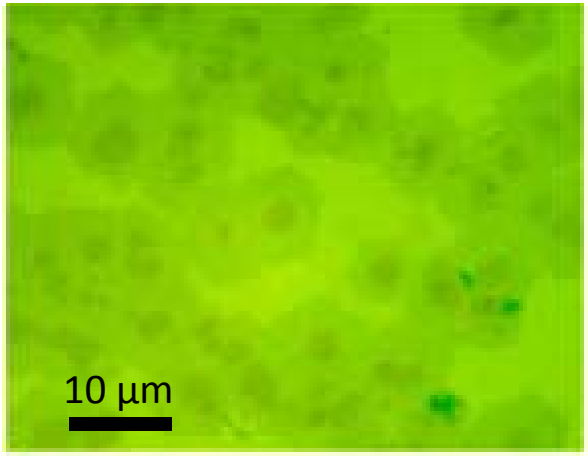
CH4: 0.1 SCCM
Growth: 21min



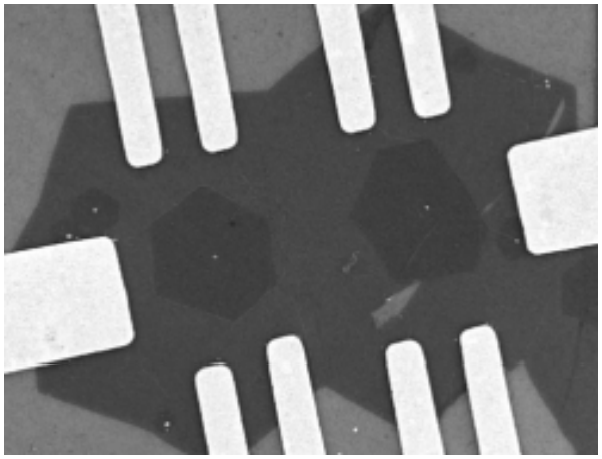
1030 – 1050 °C



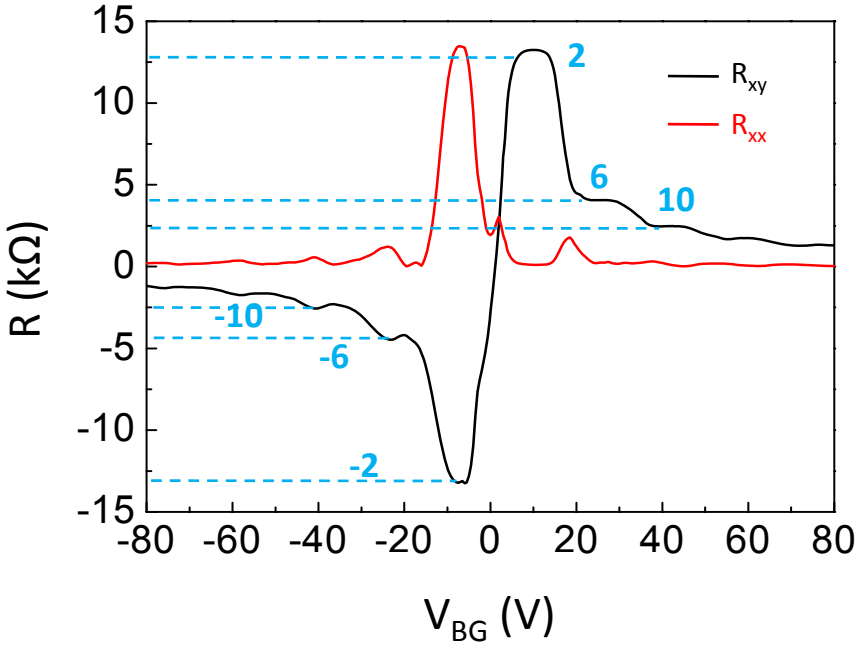
CH4: 1 SCCM
Growth: 5min

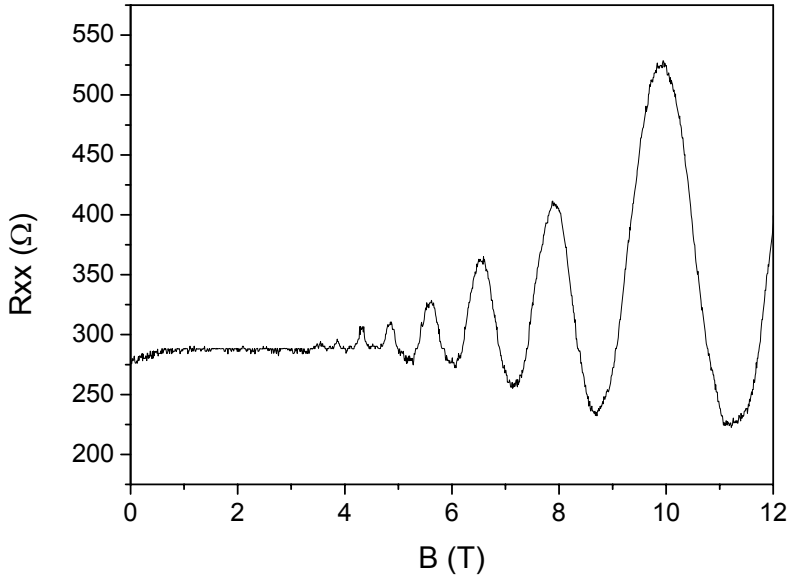


Transport properties



Half-integer quantum Hall effect for single layer



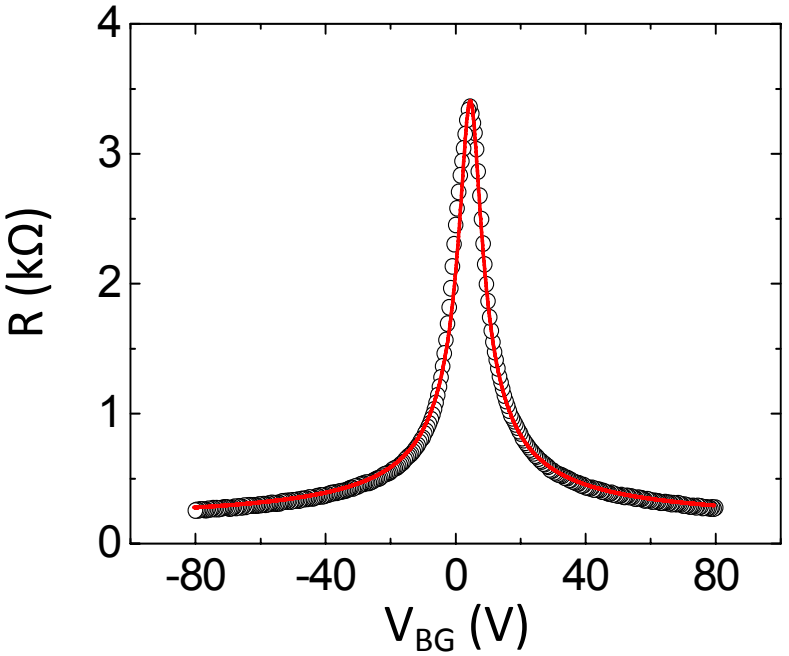


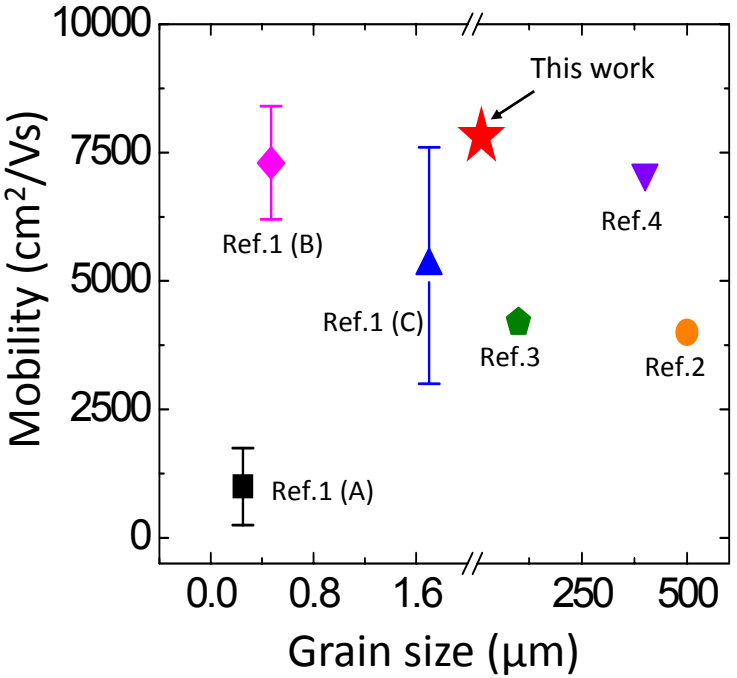
Transport properties

Extra-high mobility compared with other CVD graphene !

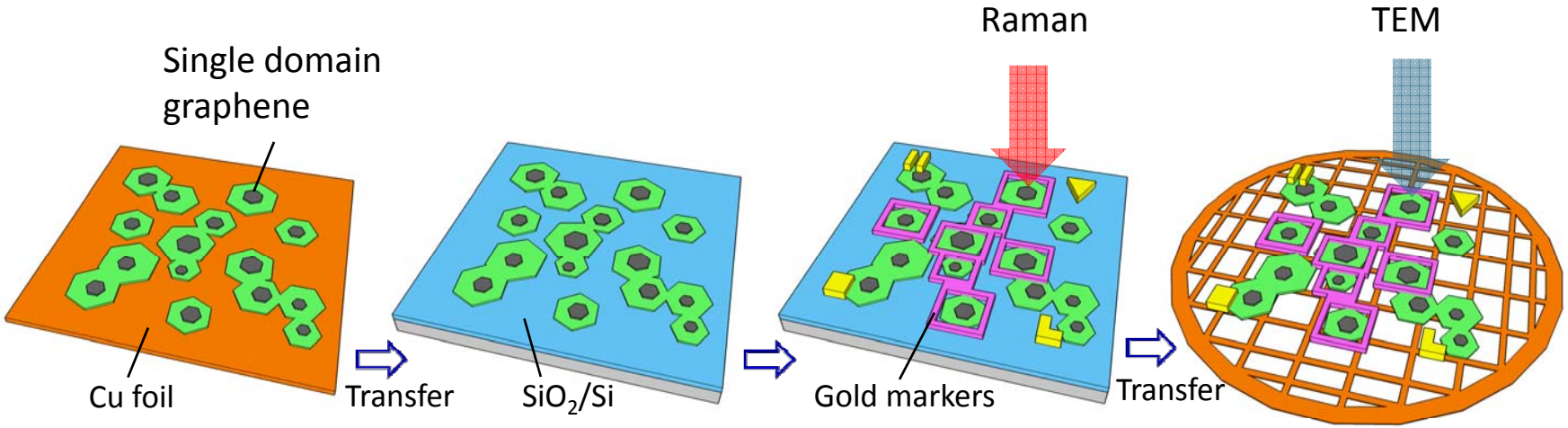
mobility = 8000 cm^2/Vs

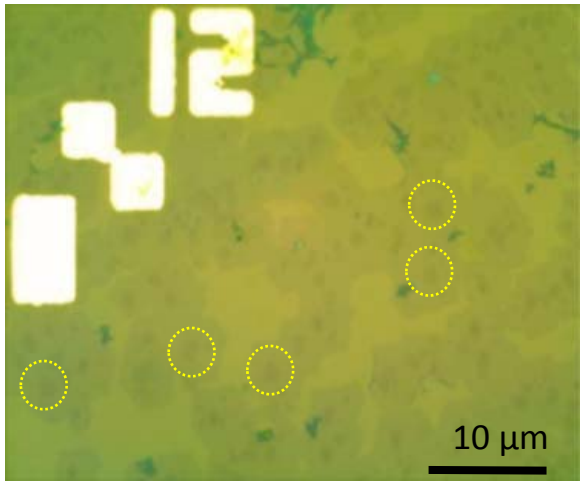
on/off ratio = 12

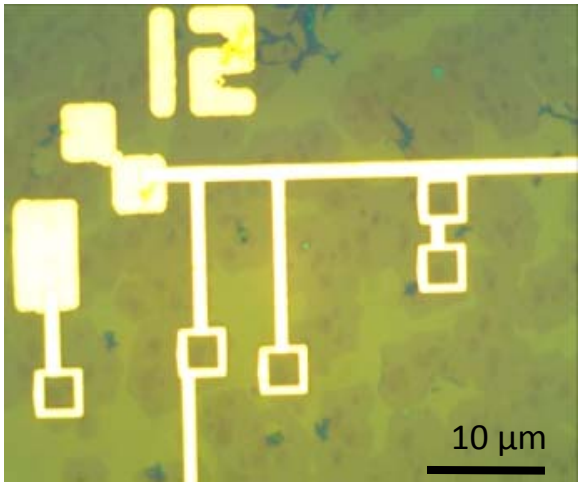


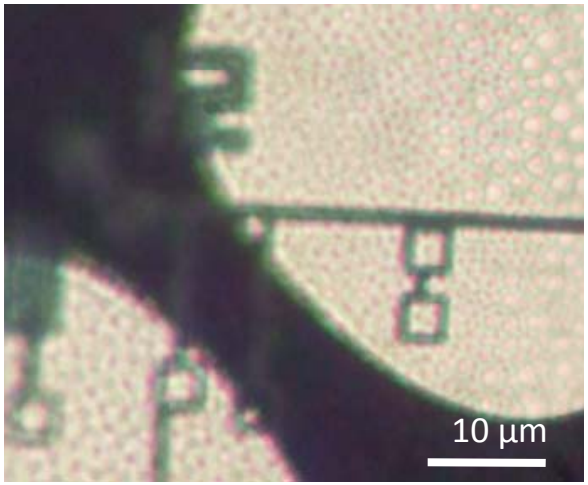


Raman + TEM

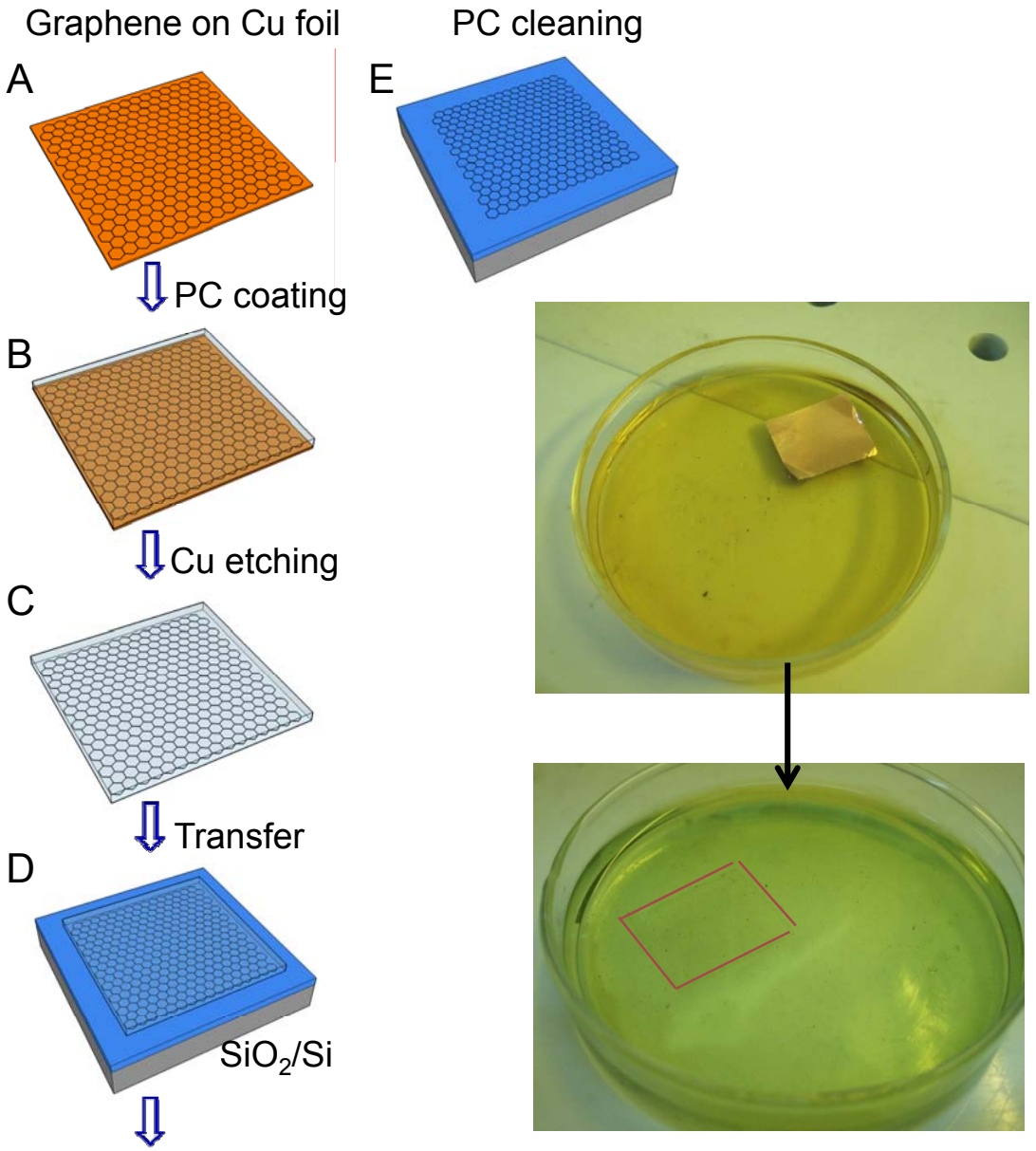

Single domain graphene
Cu foil
Transfer
SiO_2/Si
Raman
Gold markers
Transfer
TEM


10 µm


10 µm

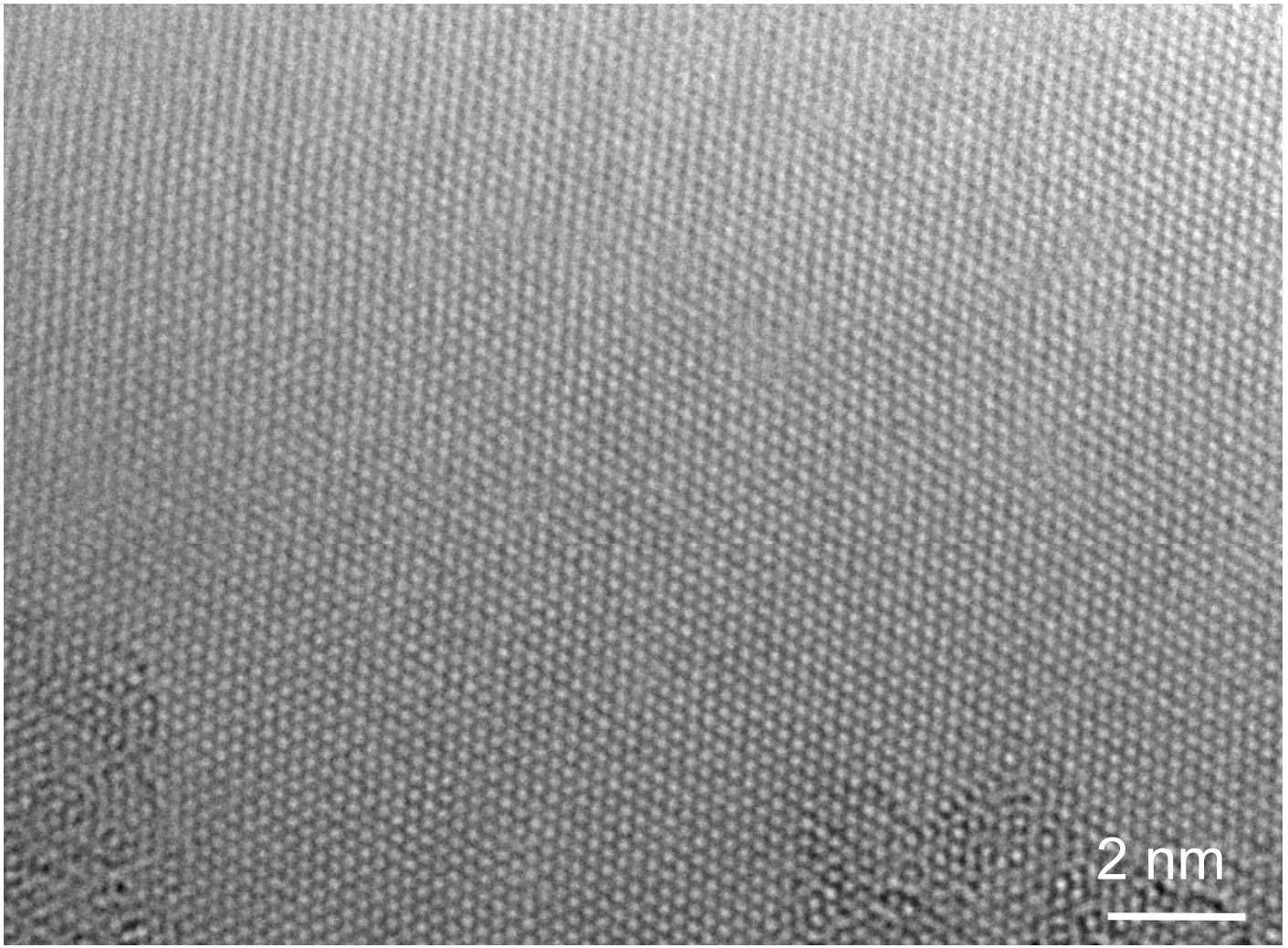

10 µm

Graphene transfer by PC



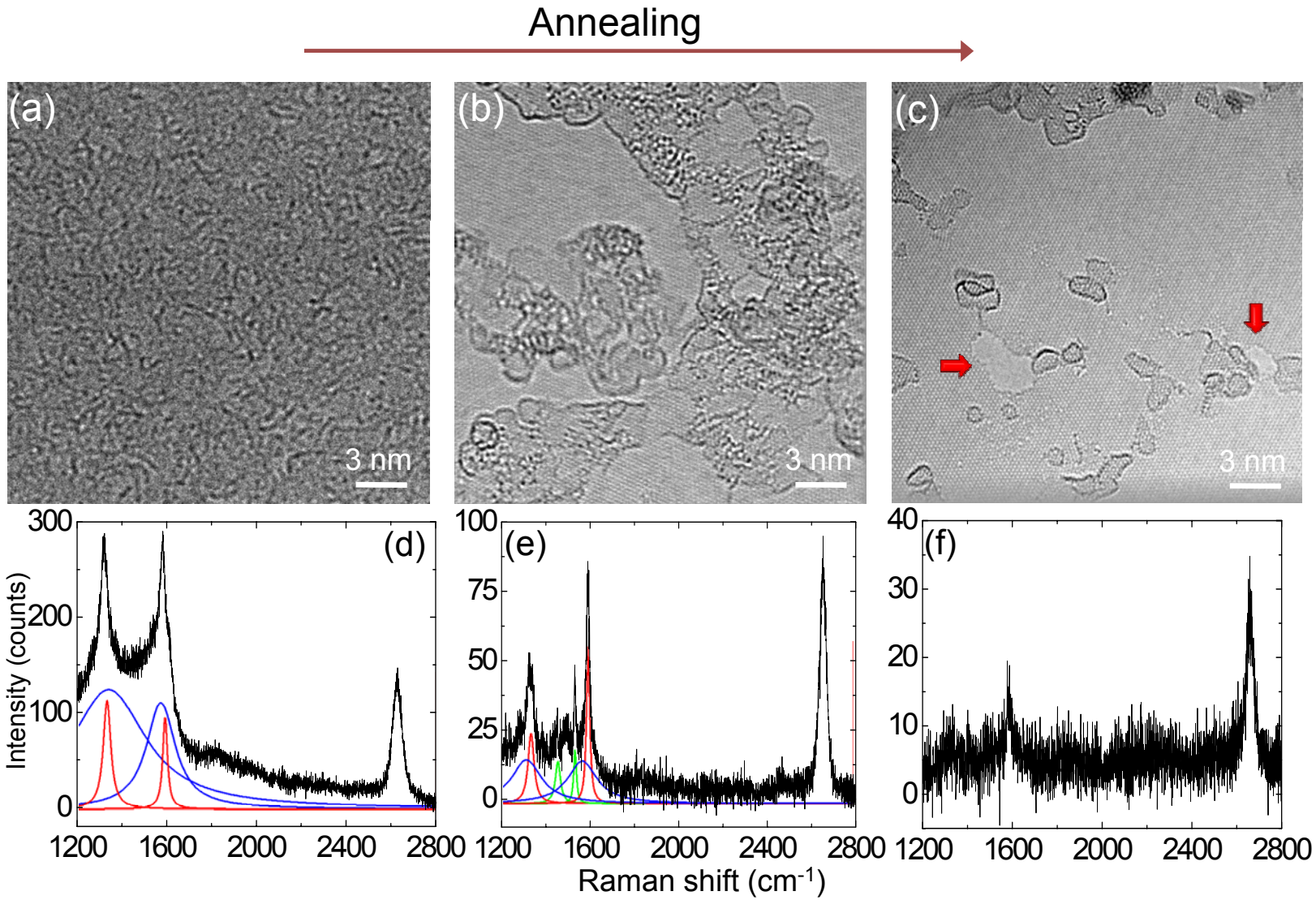
Graphene transfer by PC

HR-TEM (done in AIST)



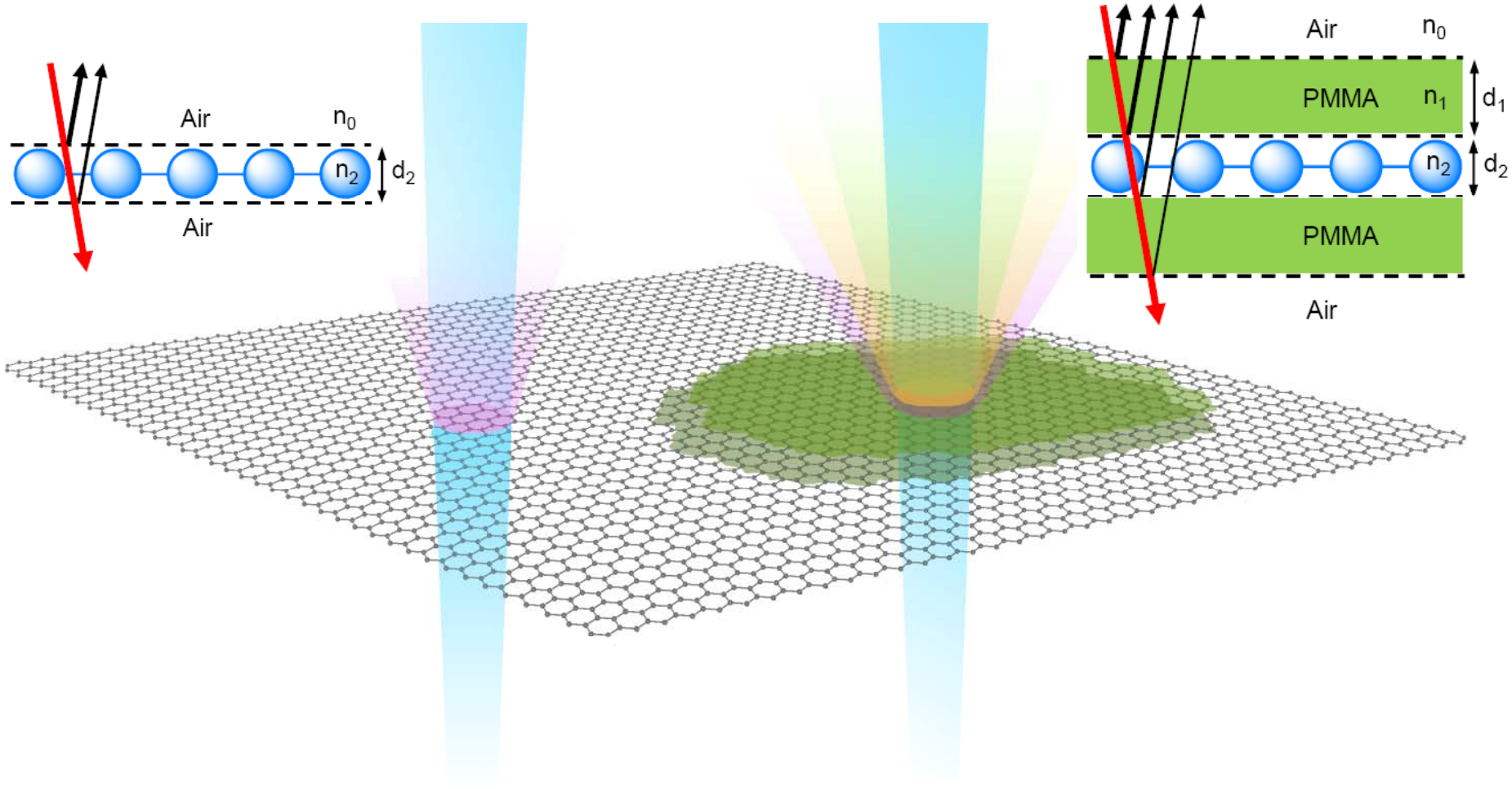
Graphene transfer by PMMA

Always a thin (1-2 nm) layer of PMMA residue on graphene !



Y. C. Lin, *et al*., ACS nano, 5, 2362 (2011).

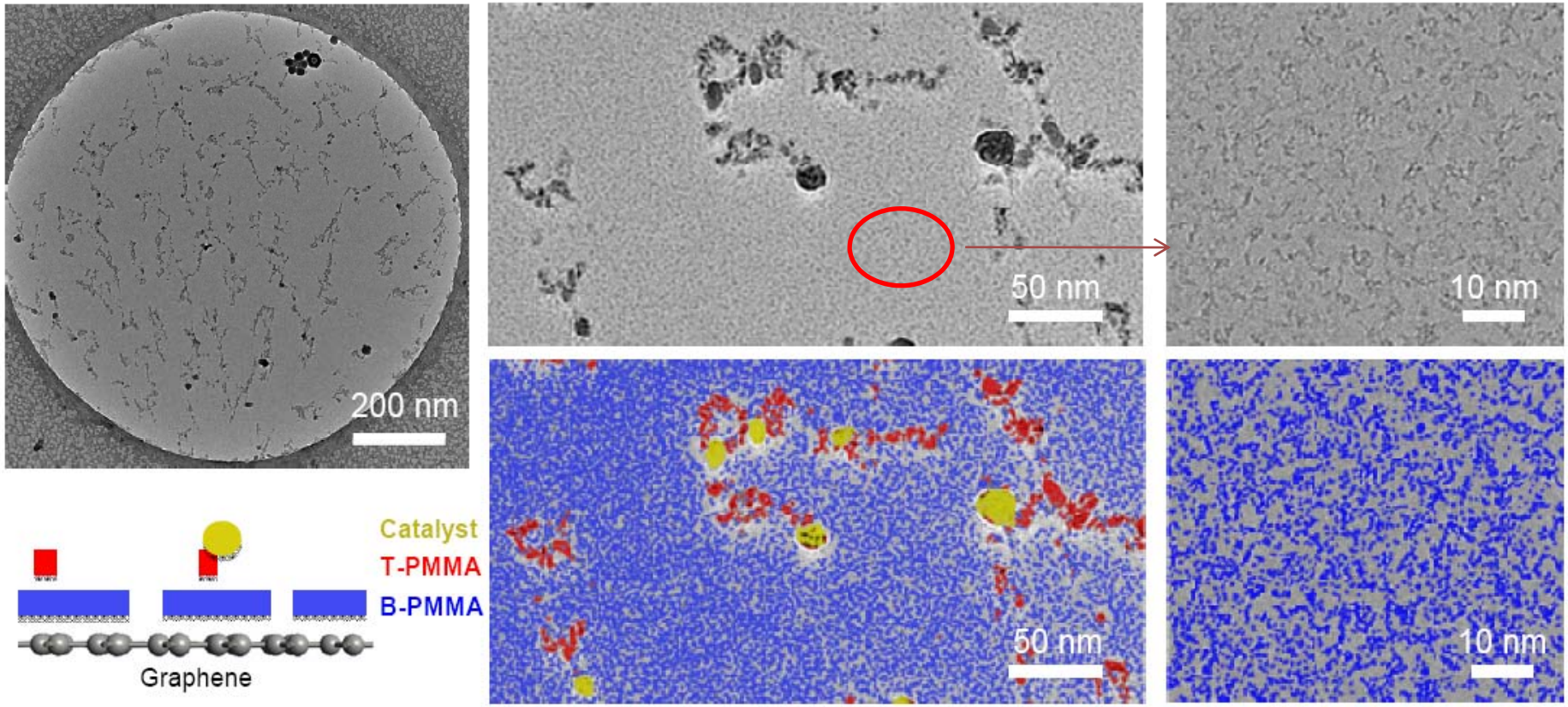
Graphene transfer by PMMA



Y. C. Lin, *et al*., ACS nano, 5, 2362 (2011)

Mobility reduction due to annealing

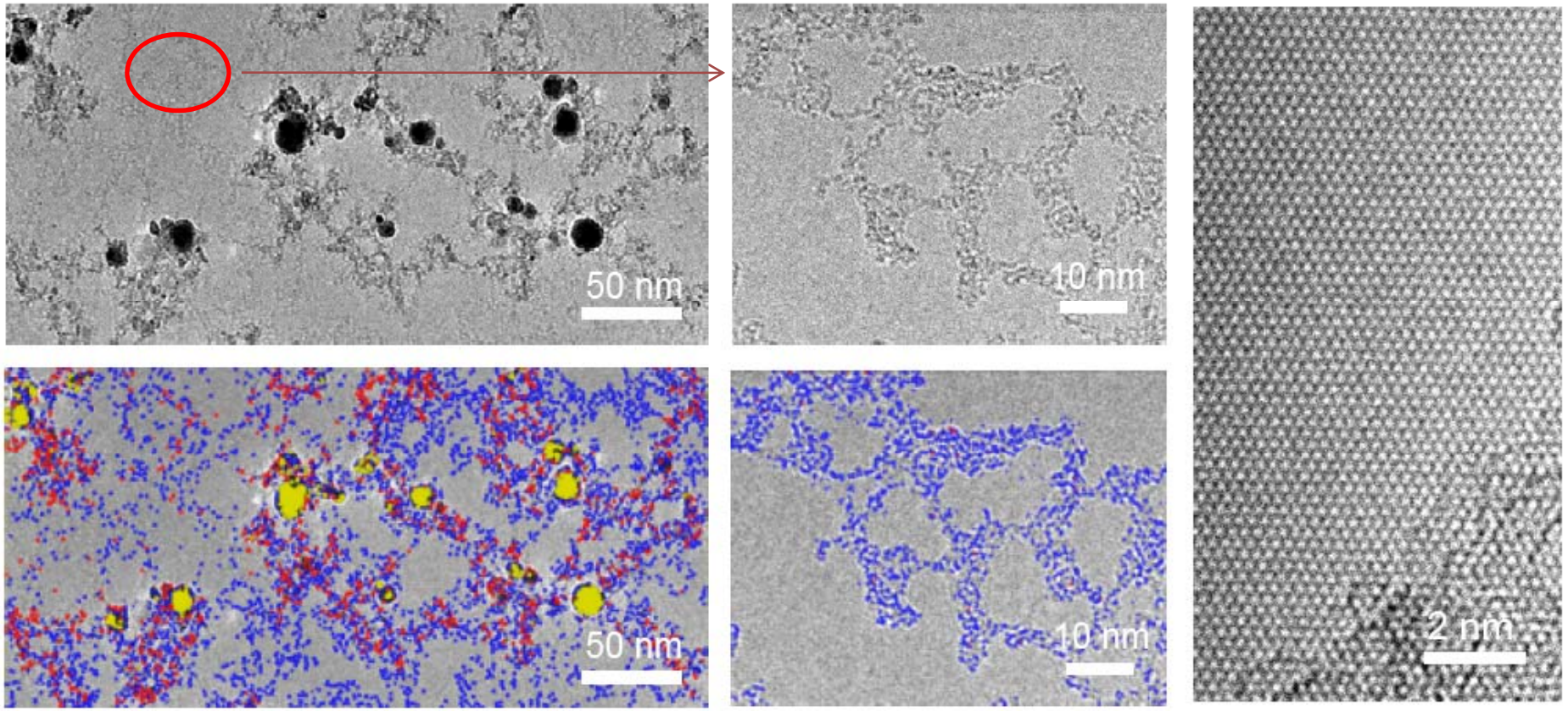
TEM images of graphene annealing at 200 °C



Y. C. Lin, *et al*., Nano Lett., 12, 414 (2012)

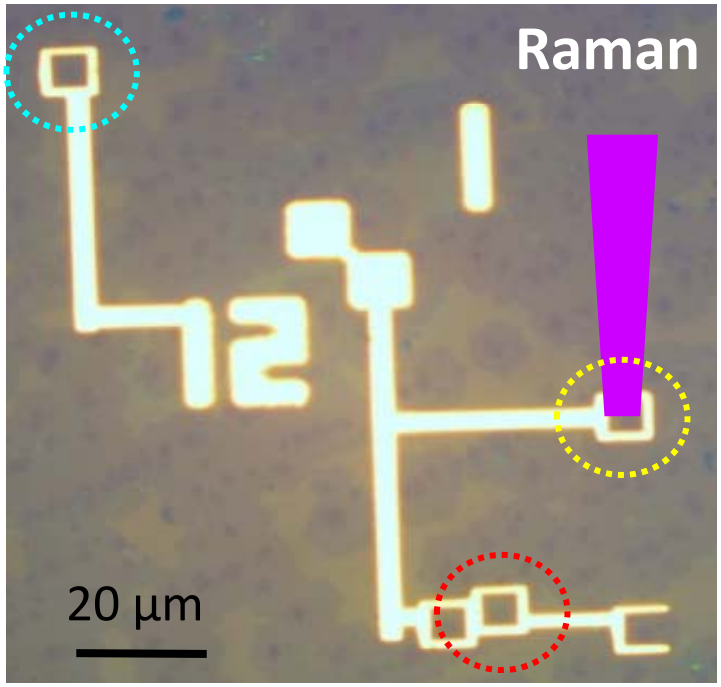
Mobility reduction due to annealing

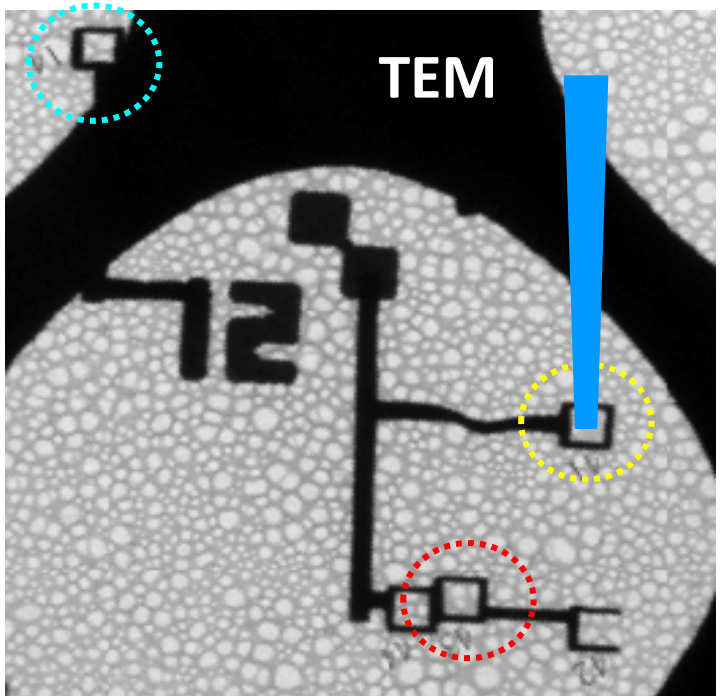
TEM images of graphene annealing at 250 °C



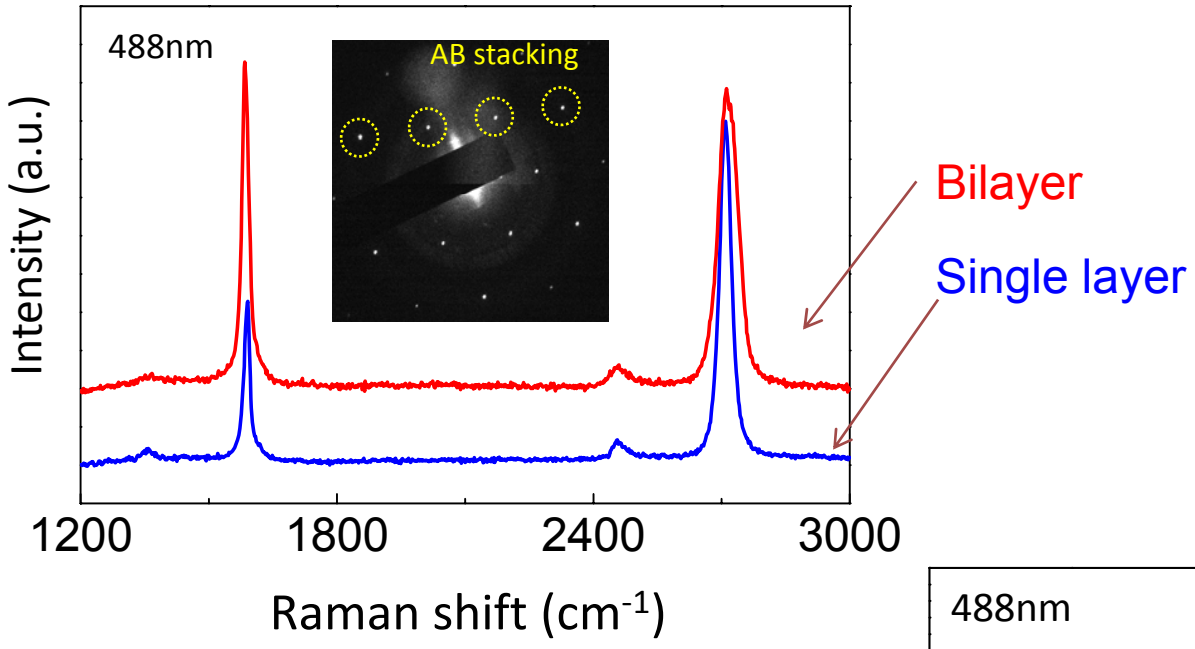
Y. C. Lin, *et al*., Nano Lett., 12, 414 (2012)

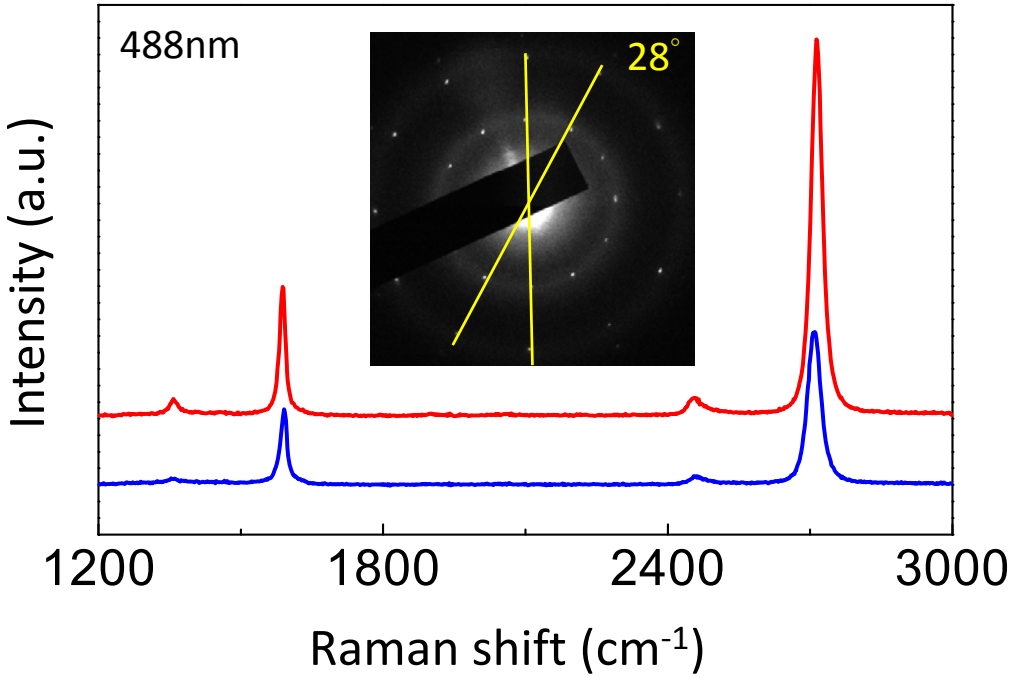
Raman + TEM





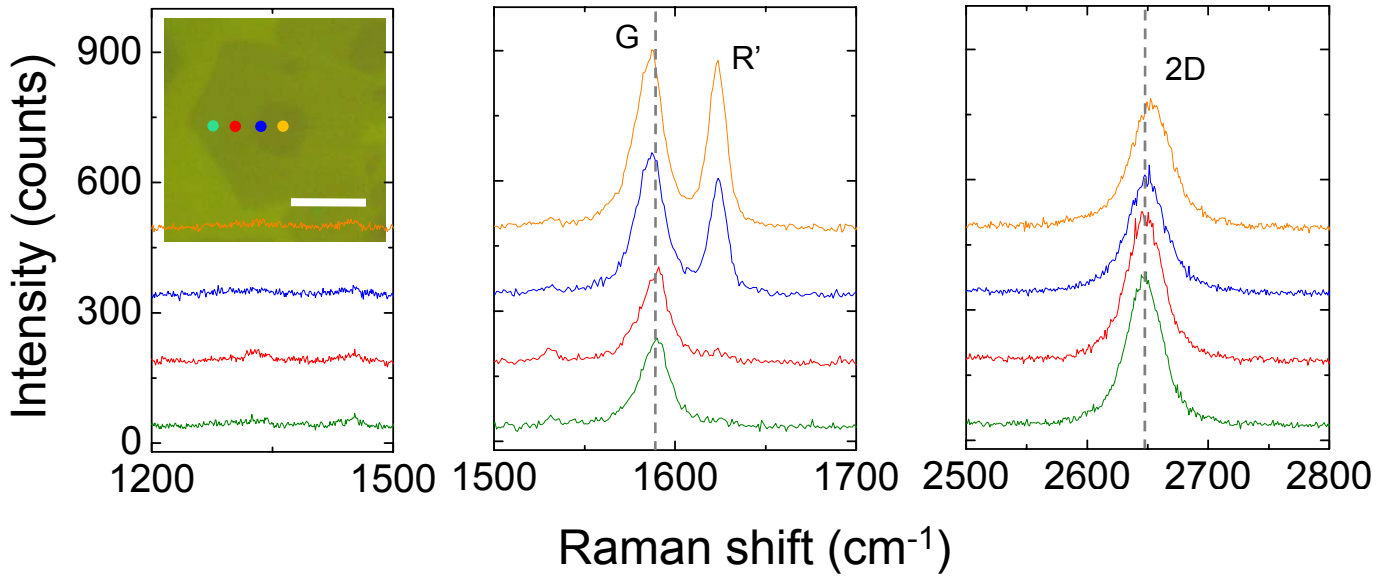
Angle-dependent Raman spectra





Angle-dependent Raman spectra

No.12_10_47 (6°)



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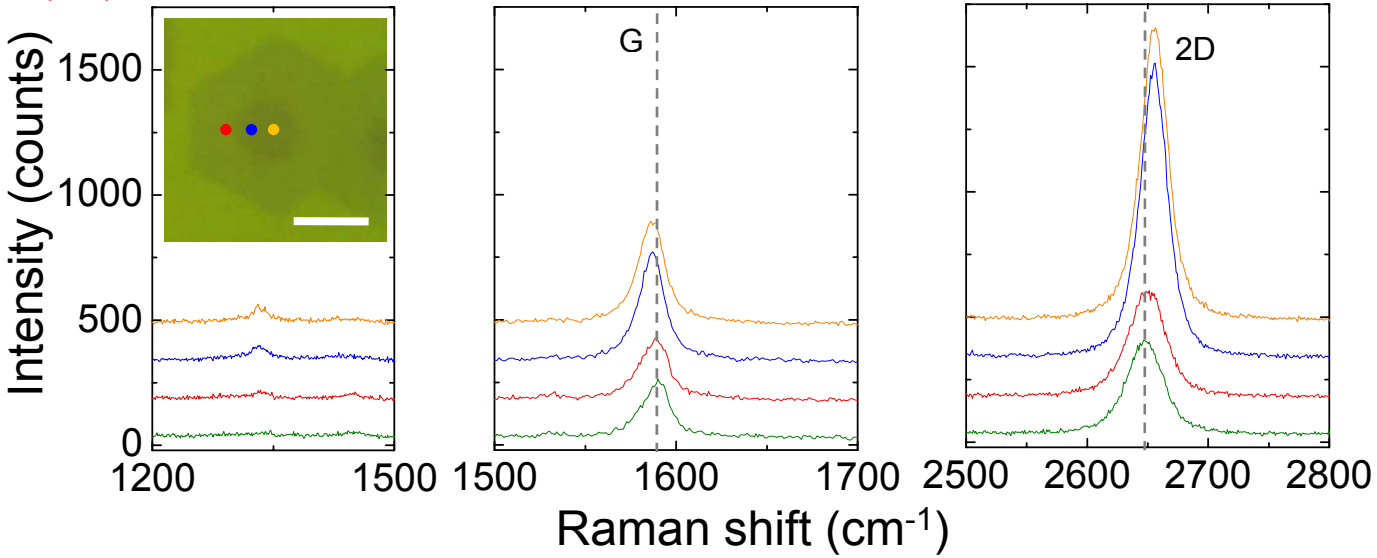
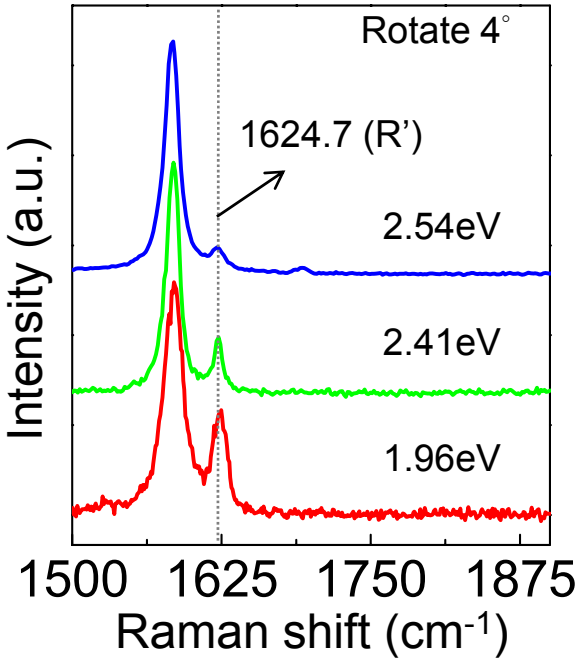
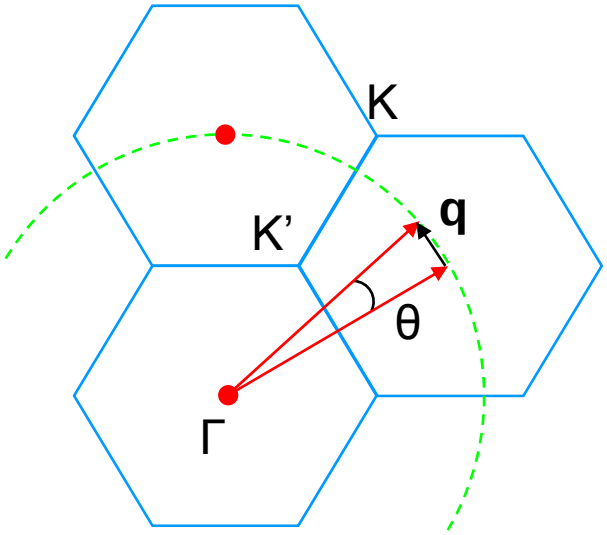


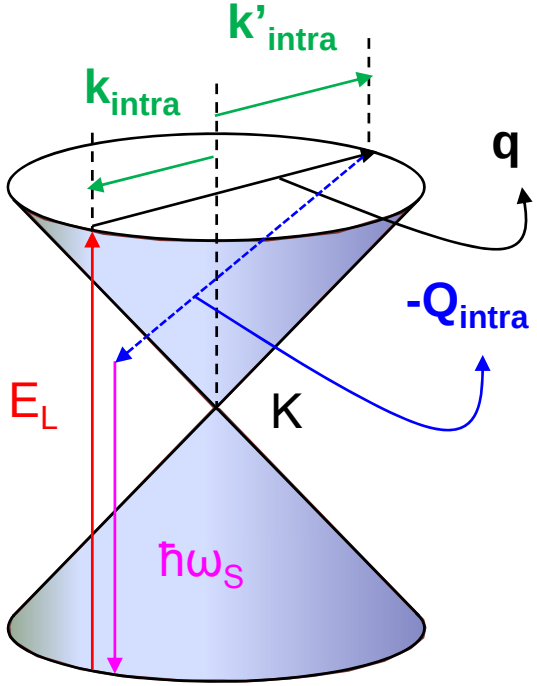
Angle dependent Raman spectra



New Raman feature: R' mode

1. non-dispersive
2. intravalley double resonance

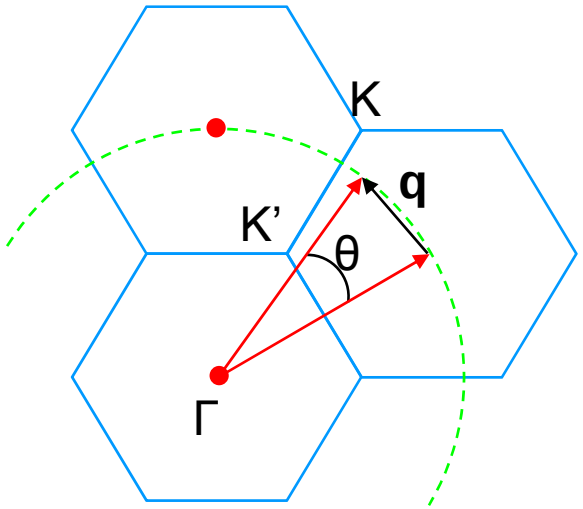


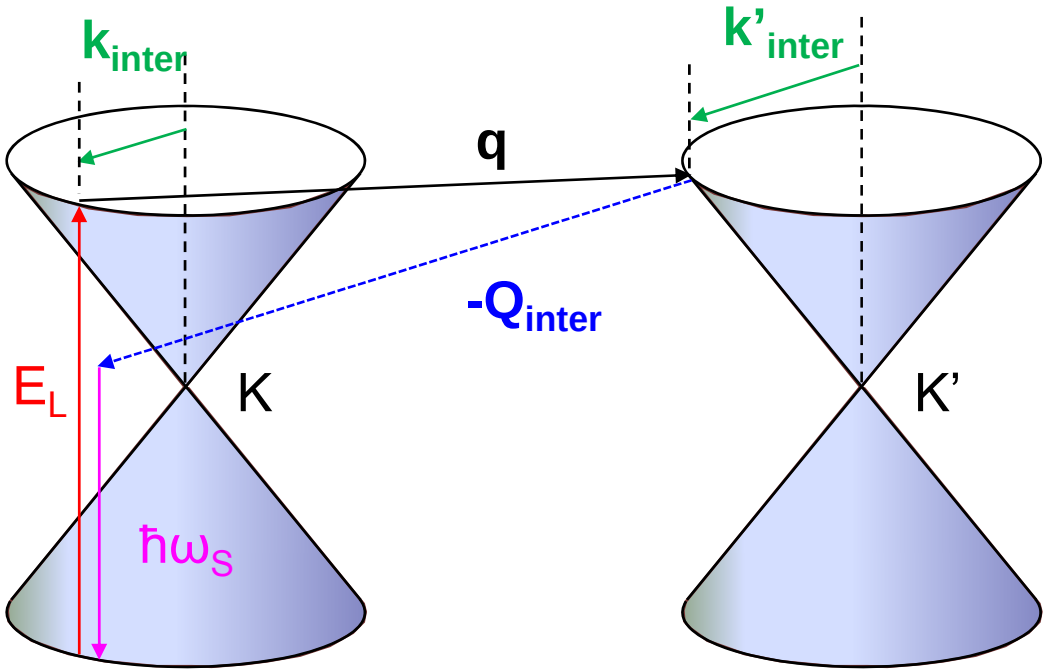


Angle-dependent Raman spectra

New Raman feature: R mode

1. non-dispersive
2. intervalley double resonance





NANO LETTERS

LETTER

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Raman Signature of Graphene Superlattices

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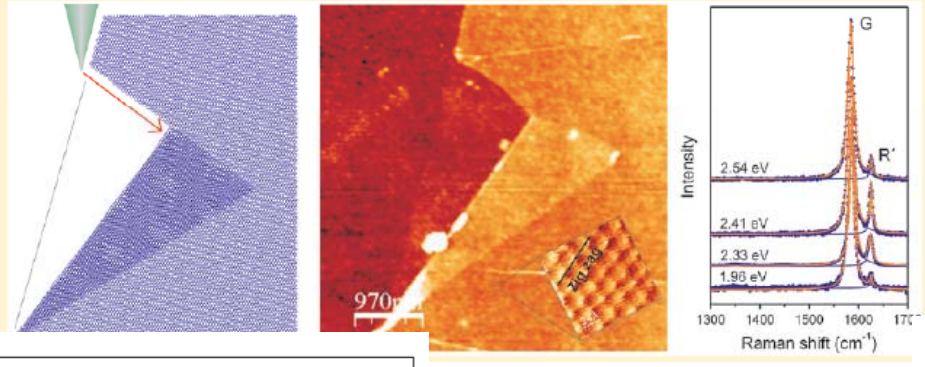
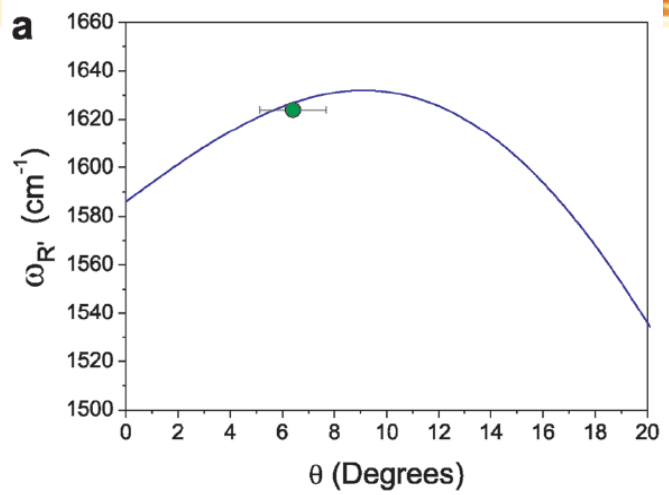
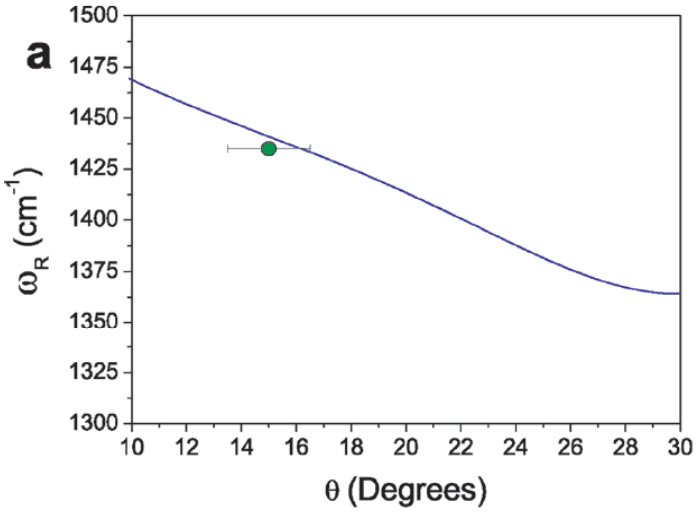
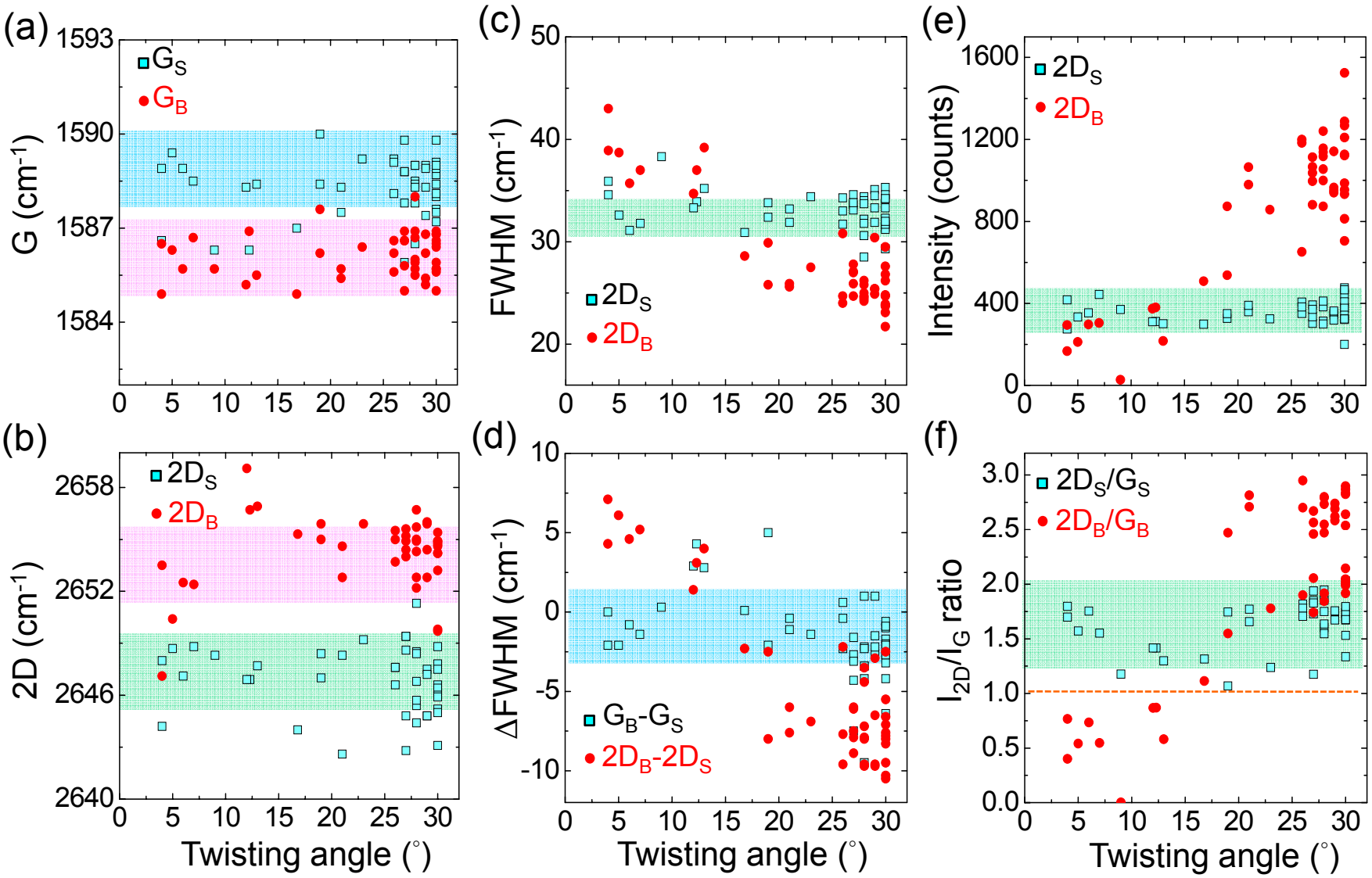
ABSTRACT:





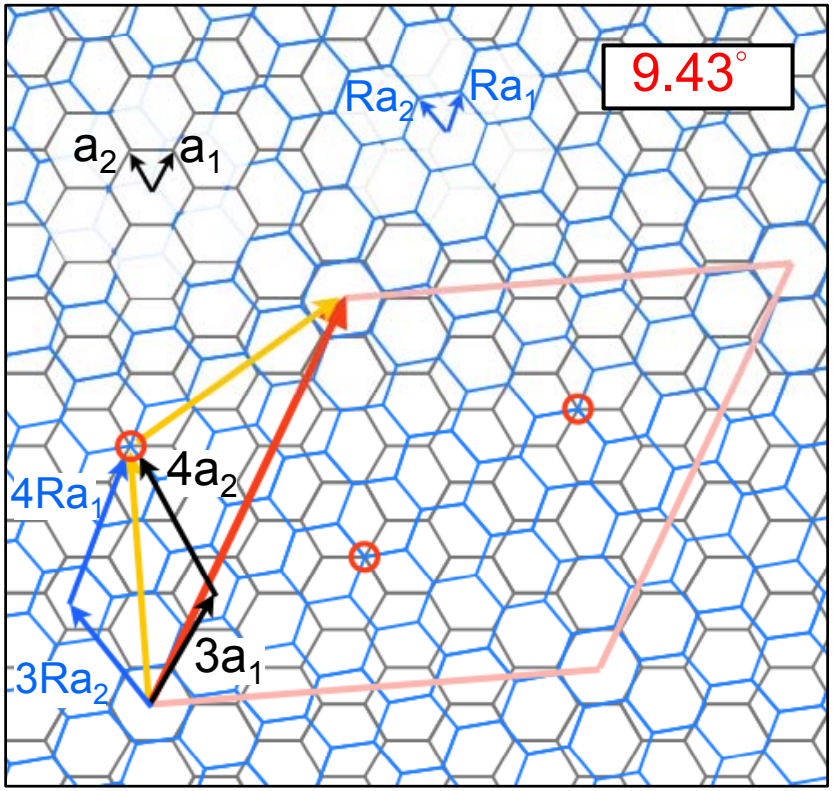


Angle-dependent Raman spectra

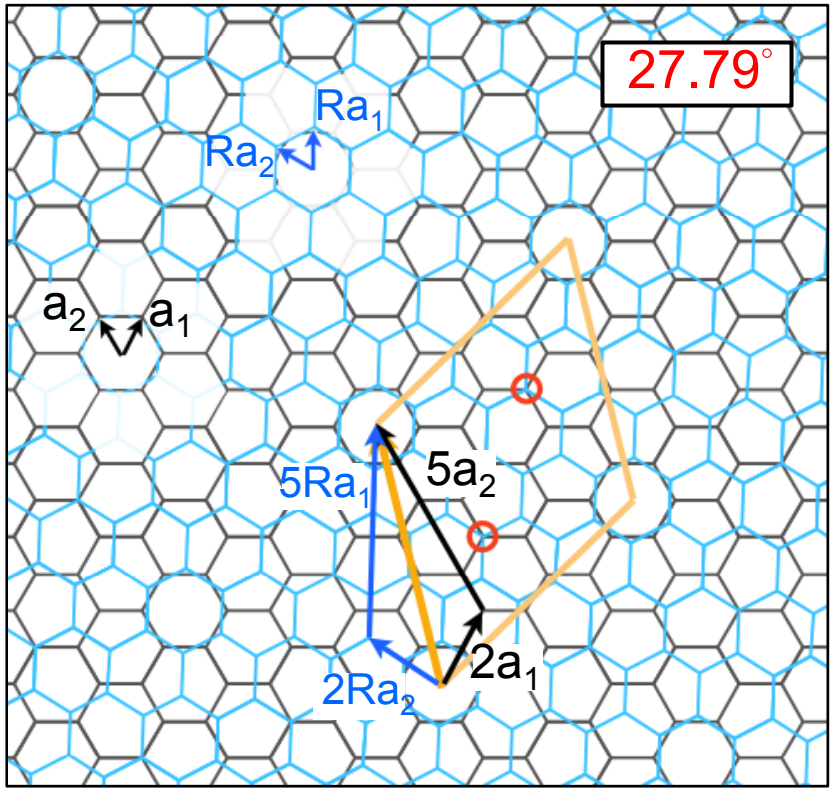


Supercell of twisted bilayer

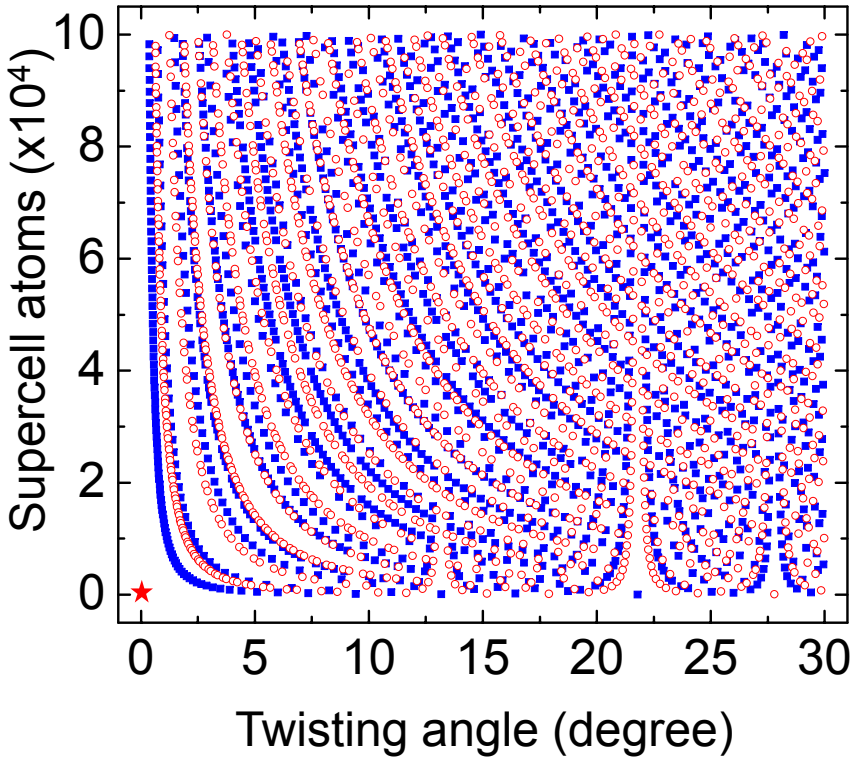
(n,m)=(3,4) K type: n-m=3i±1

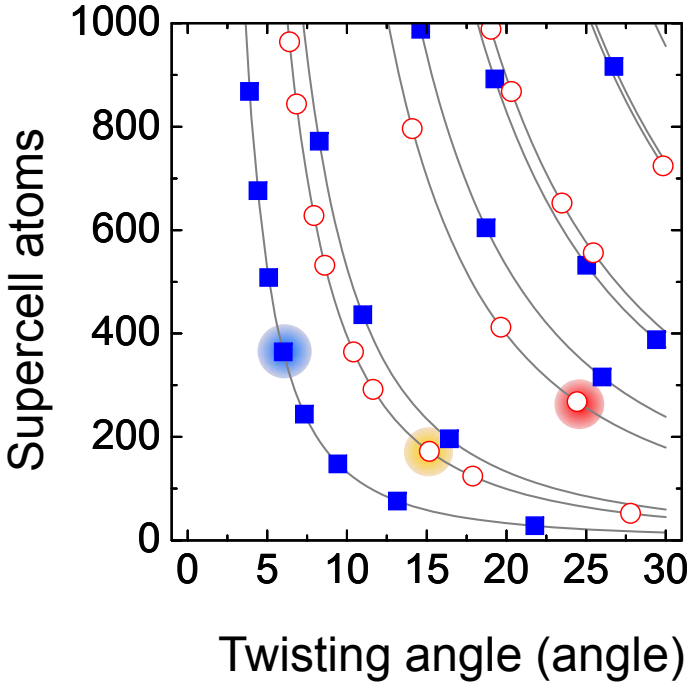


(n,m)=(5,2) Γ type: n-m=3i

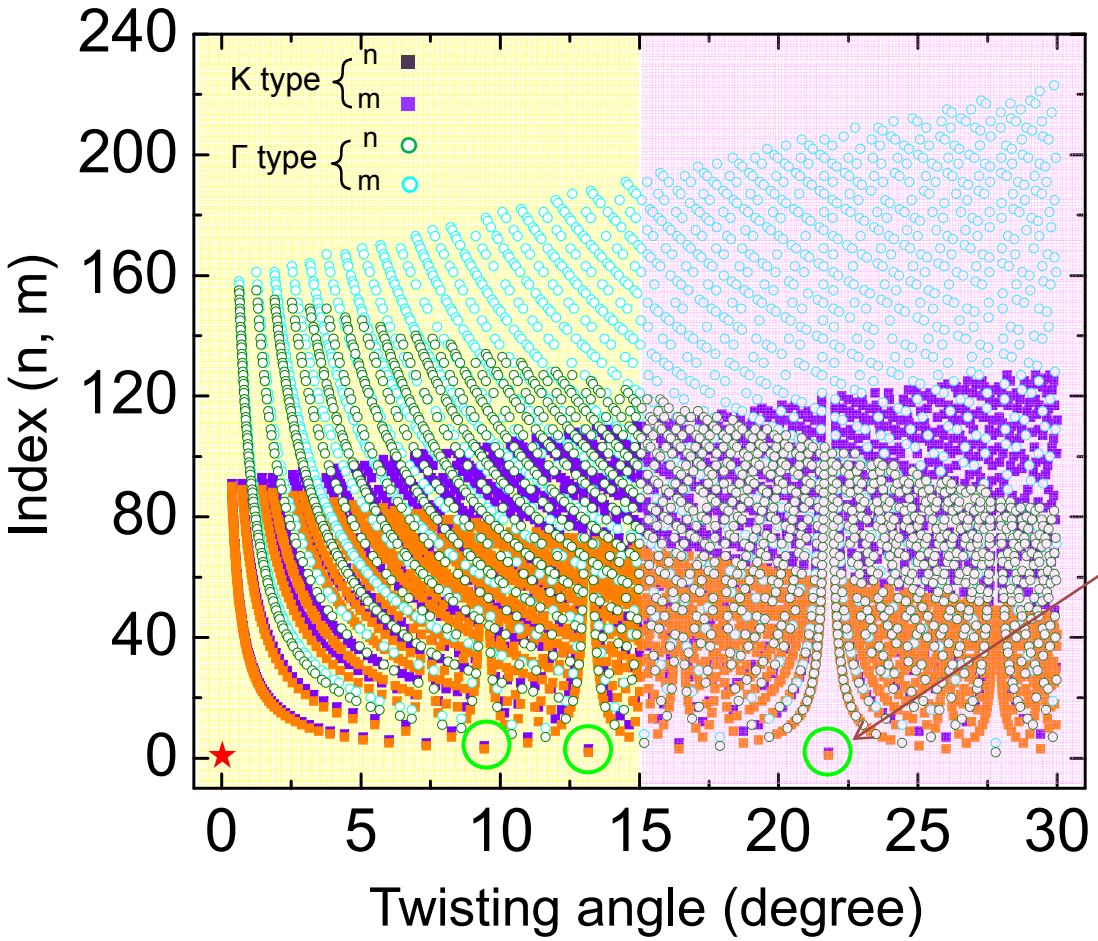


Supercell of twisted bilayer

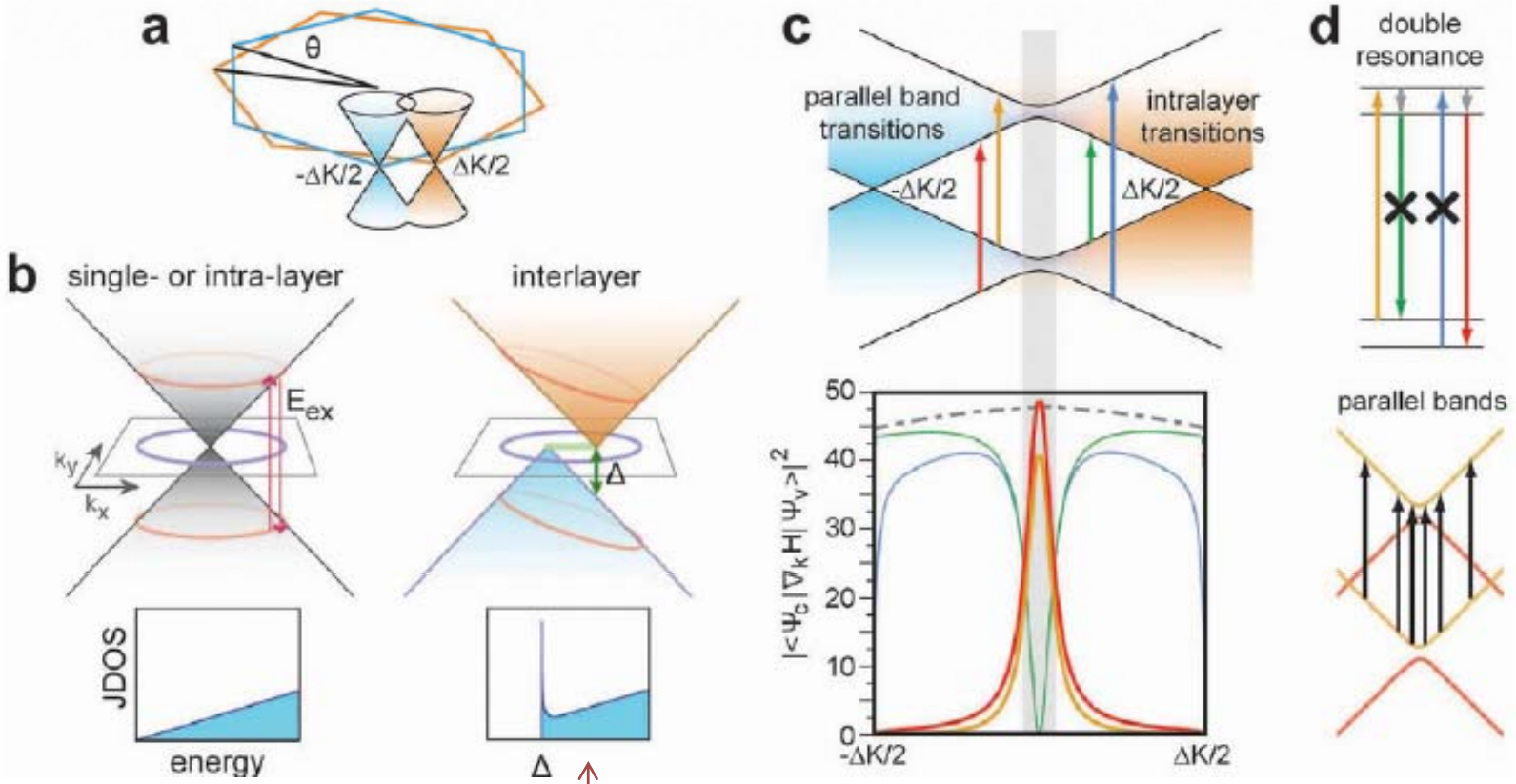


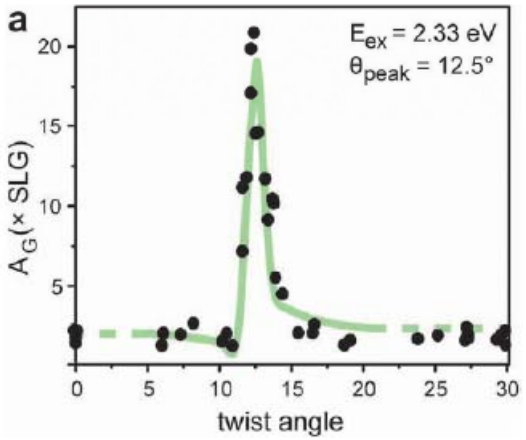


Supercell of twisted bilayer



Coupling or decoupling?



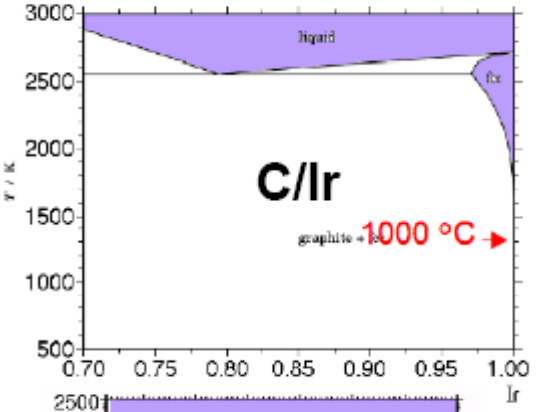


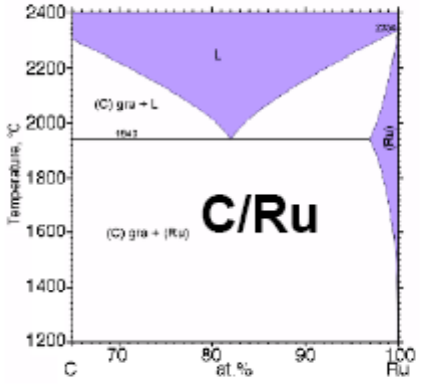
G peak enhancement due to the low-energy singularity

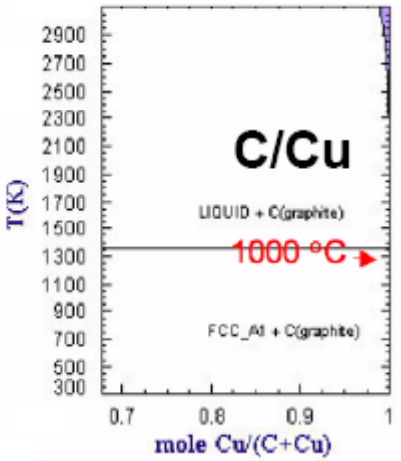
Coupling or decoupling?

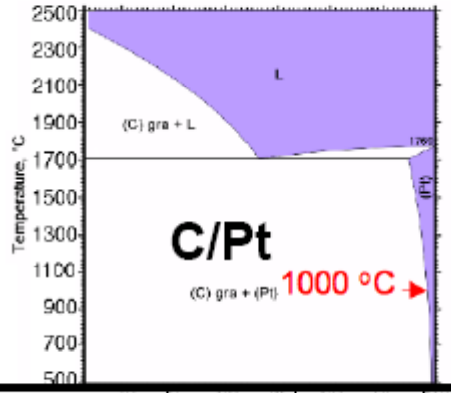
Thank you!

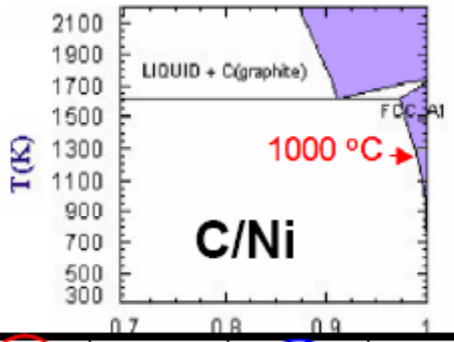
Graphene Growth on Metal Substrates

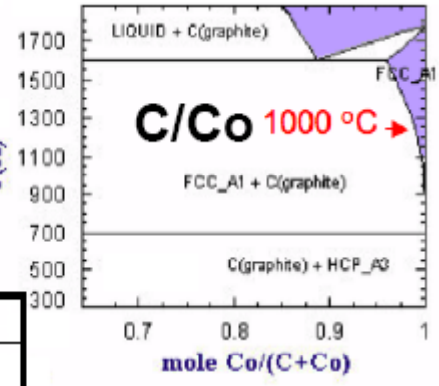












C	Ir	Ru	Cu	Pt	Ni	Co
C solubility	Low	Low	Low	High	High	High
Transfer	No	No	Yes	No	Yes	Yes
Cost	High	High	Low	High	Low	High