SAND2010-7375C

Characterization of devices fabricated from electrostatically transferred graphene: comparison with epitaxial based devices

Stephen W. Howell

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Sandia National Laboratories

October 20, 2010

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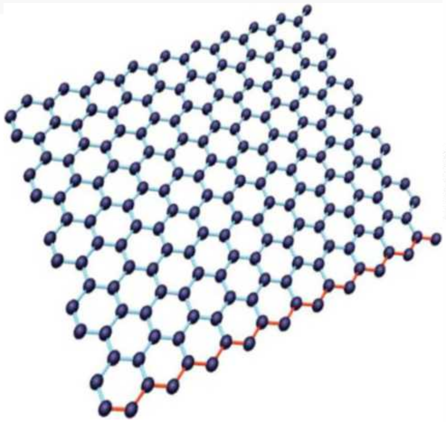
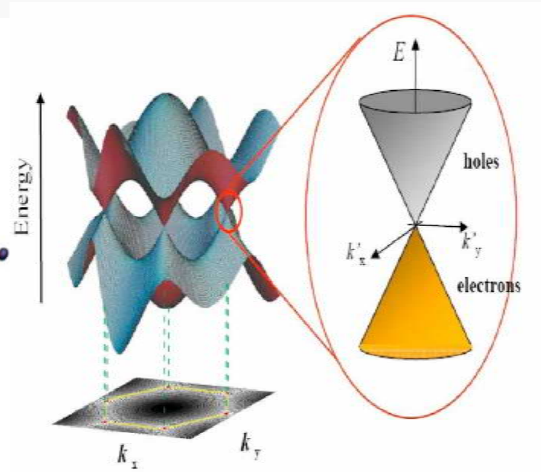
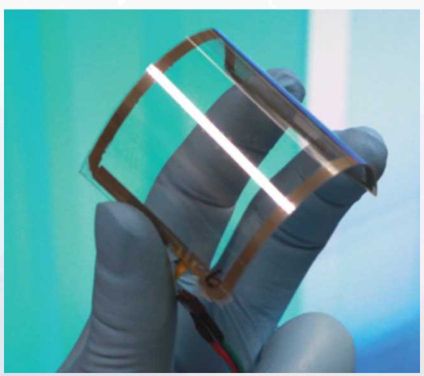
Why graphene?





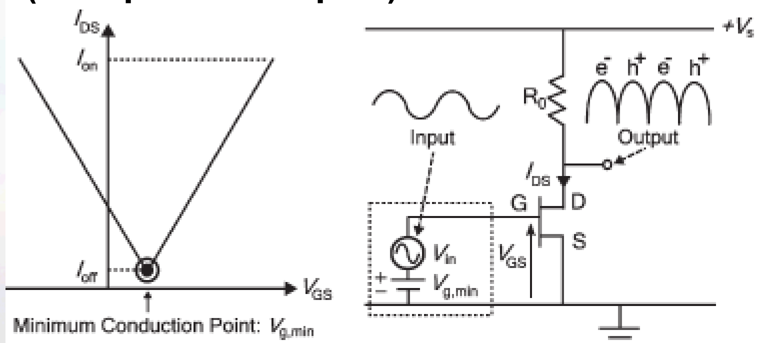
Mobility, μ, up to 250,000 cm^2/Vs
(suspended exfoliated graphene)
Ambipolar, zero-bandgap
Current densities up to 5 x 10^8 A/cm^2
Elastic modulus ~ 1 TPa

Transparent electrodes



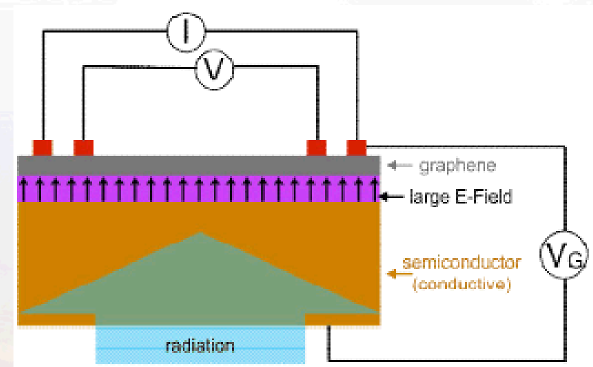
S. Bae *et al, Nature Nano.*, **5**, 574-578 (2010)

Frequency multiplier (ambipolar transport)



H. Wang *et al., IEEE Elec. Dev. Lett.*, **30**, 547-549 (2009).

Read-out for radiation detection

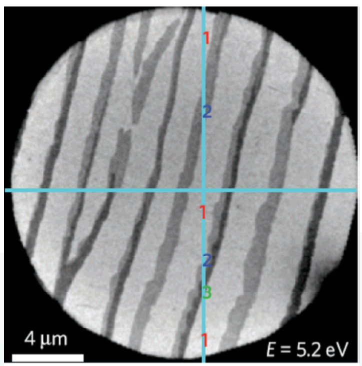


M. Foxe et al., *IEEE Trans. Nuclear Sci.*, submitted (2010).

Sandia's graphene effort

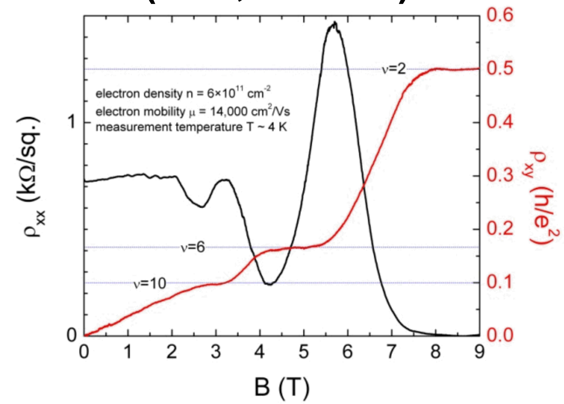
•Sandia is in the 3rd year of an internally funded project that is focused on developing the scientific biases required for synthesis of high quality graphene

Thermal Decomposition of SiC (Ohta)

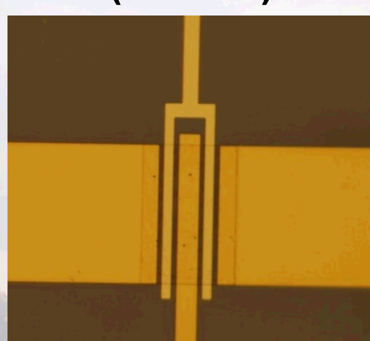


CVD Dep on Metals (McCarty)

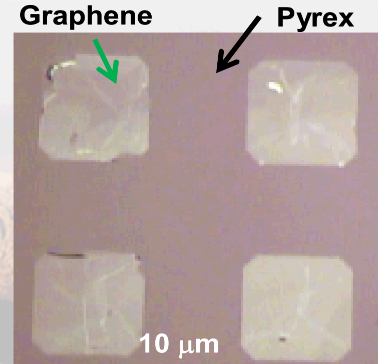
Transport Measurements (Pan, Howell)



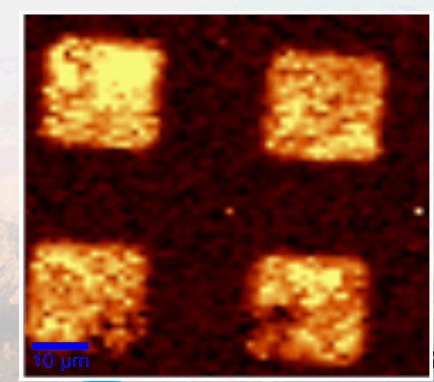
GFET Development (Howell)



Transfer (Biedermann, Howell)



Raman Mapping (Beechem, Ohta)

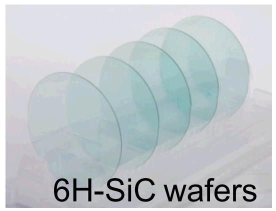


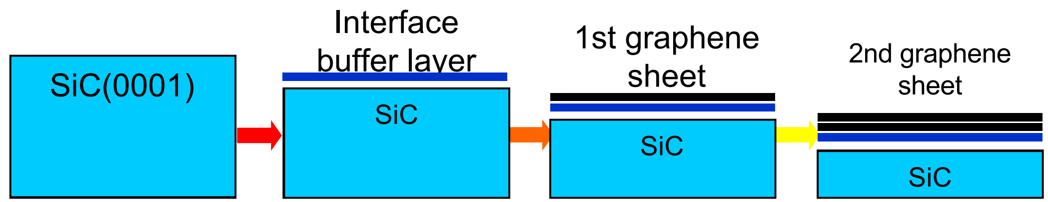
Motivation for transferring epitaxial graphene (EG) from SiC

- **Why use EG on SiC?**
  - EG grown on SiC has the potential for high carrier mobility
  - Current synthesis approaches allow for tight control of graphene thickness (monolayer and bilayer coverage)
- **Development of a scalable transfer technology could:**
  - Enable EG integration with current CMOS processes
  - Enable new types of MEMS device structures

Synthesis approach for epitaxial graphene (EG) on SiC

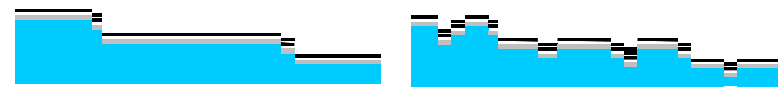
- **Graphitization of SiC:**
  - **Sublimation of Si at high temperature (>1200 °C) leaves graphene layer at SiC surface**

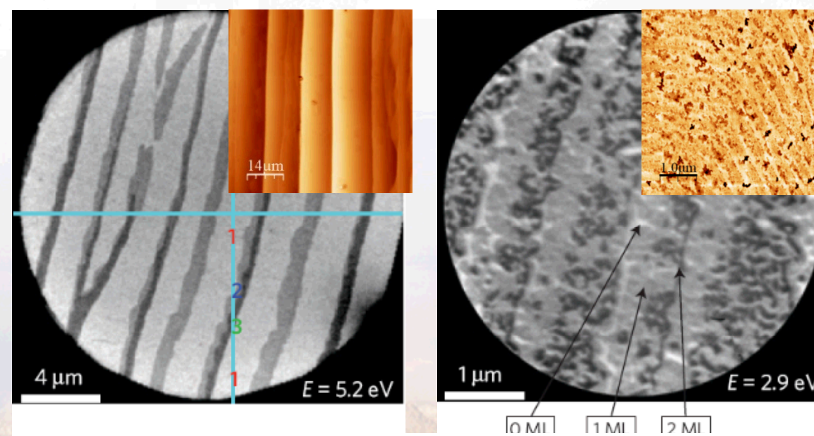




Atmospheric pressure Ar high temp. processing

Ultrahigh vacuum mid temp. processing





•Argon-assisted graphene formation yields large (~100 μm²) graphene domains

•Samples prepared using Ar atmosphere at atmospheric pressure and high temp

•Using this method we have exquisite control of :
- **•Domain size**
- **•Percentage coverage of mono/bilayers**

K. V. Emtsev et al., Nature Mater. 8, 203 (2009).
C. Virojanadara et al., Phys. Rev. B 78, 245403 (2008).

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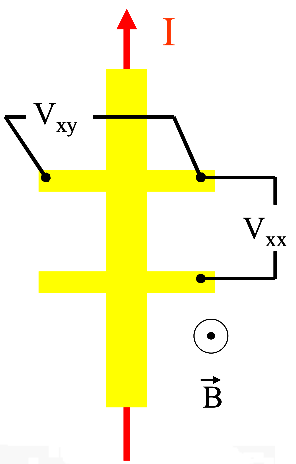
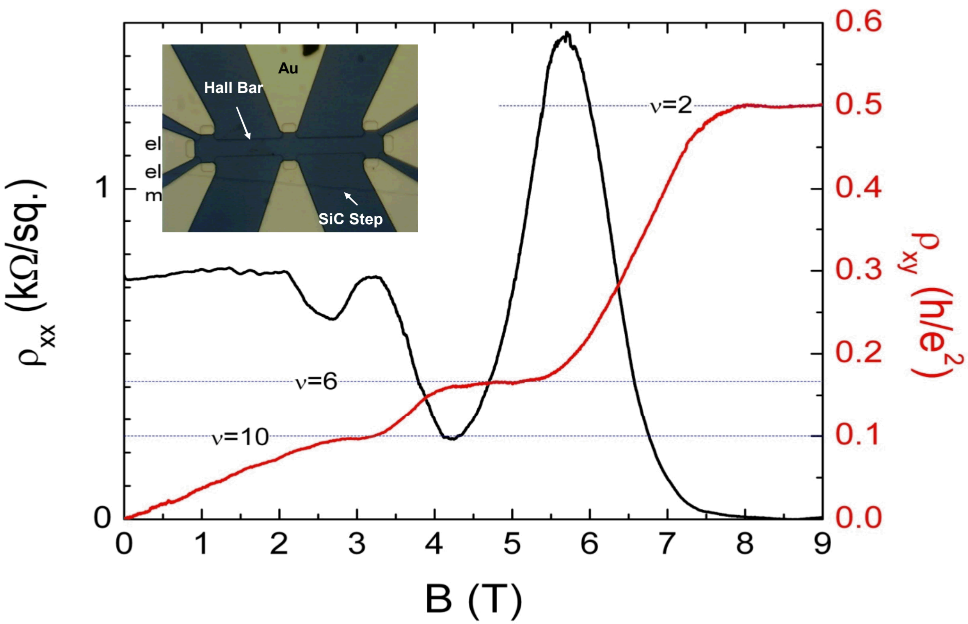
Electronic characterization of EG grown on SiC

Low Temp Transport Measurements

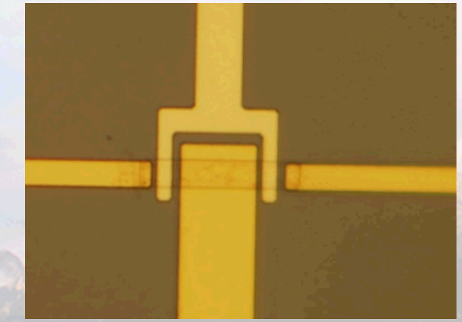


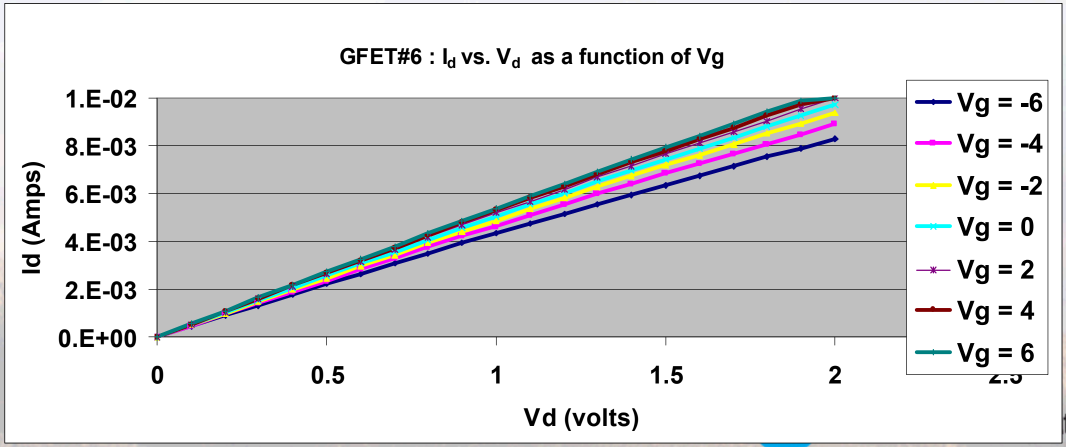


- Epi-graphene electron mobility: 14,000 cm^2/Vs
- Electron density: 6 x 1011 cm^2
- Graphene sheet resistance: ~1600 Ω/sq (average from 12 devices)
- Observed IOHE on 3 devices on the same chip

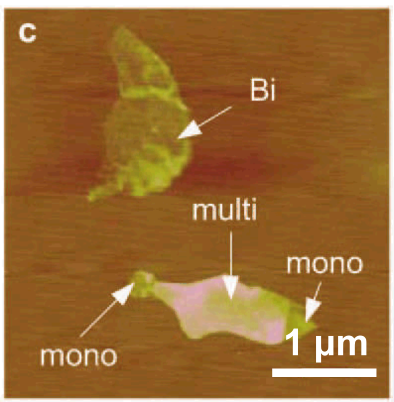
Pan *et al.*, accepted by APL

GFET Development





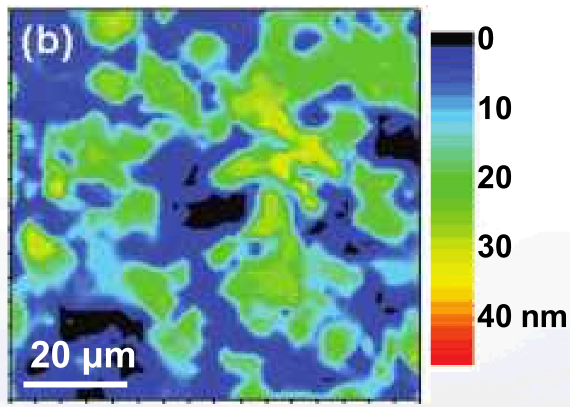
Methods to transfer EG to SiO_2



Si-face EG
Small flakes

D. Lee *et al., Nano Lett.* **8**, 4320-5 (2008).

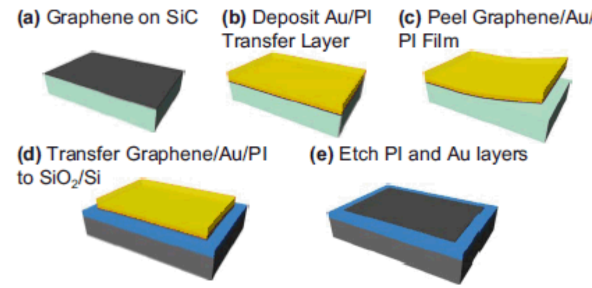
Thermal release tape
+ 5 N/mm² pressure

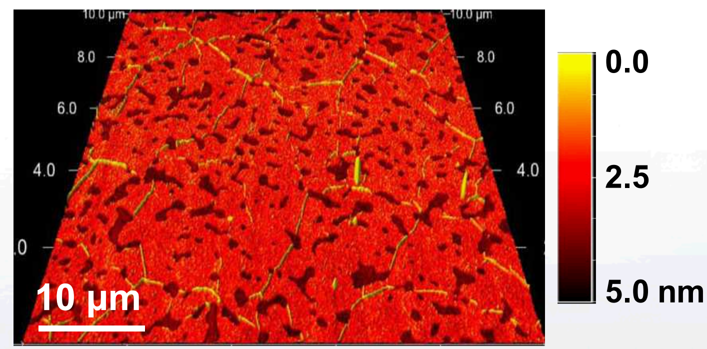


C-face EG
Very thick graphene
µ ~ 1350 cm²/Vs

J. Caldwell *et al., ACS Nano.* **4**, 118-14 (2010).

Gold/polyimide film handle





Si-face EG
Damaged graphene
µ ~ 100 cm²/Vs

S. Unarunoai *et al., APL.* **95**, 202101 (2009).

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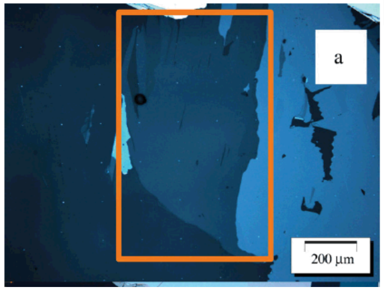
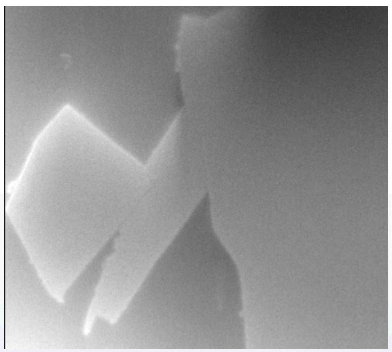
Voltage-driven exfoliation of graphite



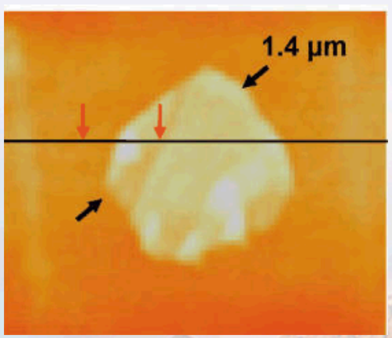
Graphite to Pyrex
1.2 – 1.7 kV
μ ~ 10^4 cm^2/Vs

A. Shukla *et al.*, *Solid State Comm.* **149**, 718-21 (2009).



HOPG to SiO_2
3-30 kV

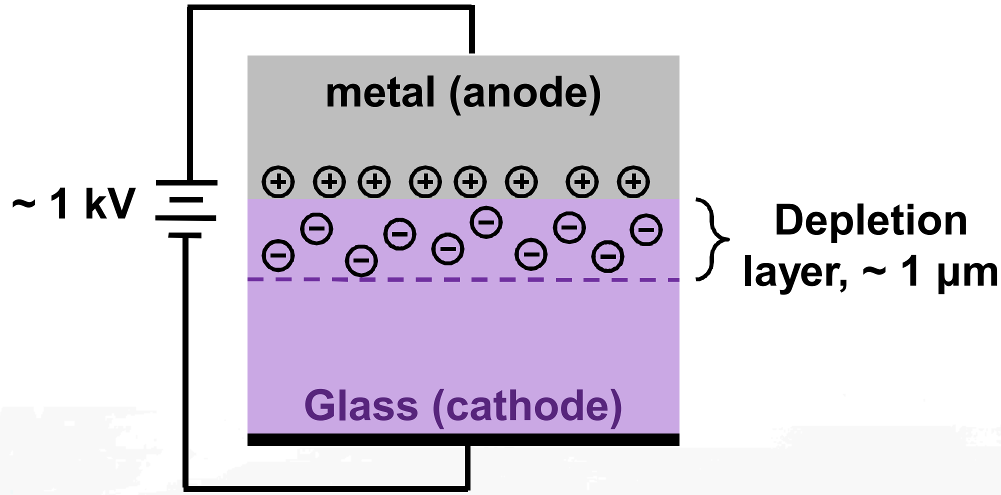
A. Sidorov *et al.*, *Nanotechnology* **18**, 135301 (2007).



Pre-patterned HOPG to SiO_2
8.5 V
μ ~ 1050 cm^2/Vs

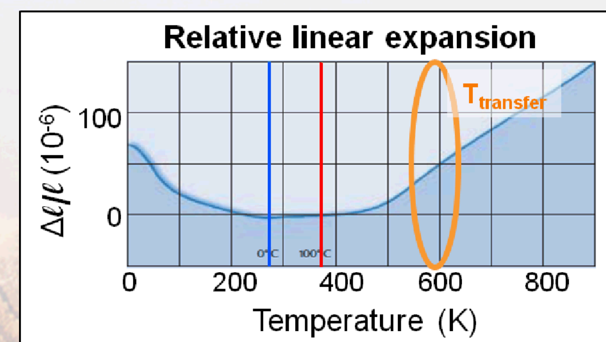
X. Liang *et al.*, *Nano Lett.* **9**, 467-72 (2009).

Shukla's method is derived from anodic bonding.



Common glasses are Pyrex, borosilicate glass, and Zerodur.

Zerodur is a Li_2O_3-Al_2O_3-SiO_2 glass ceramic



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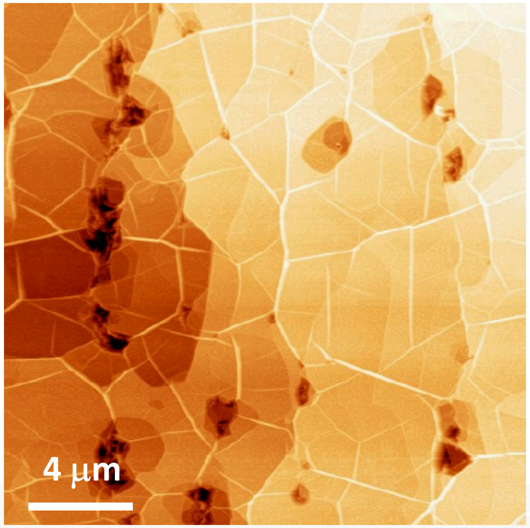
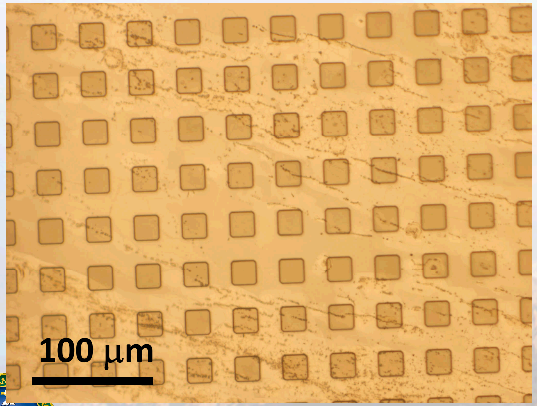
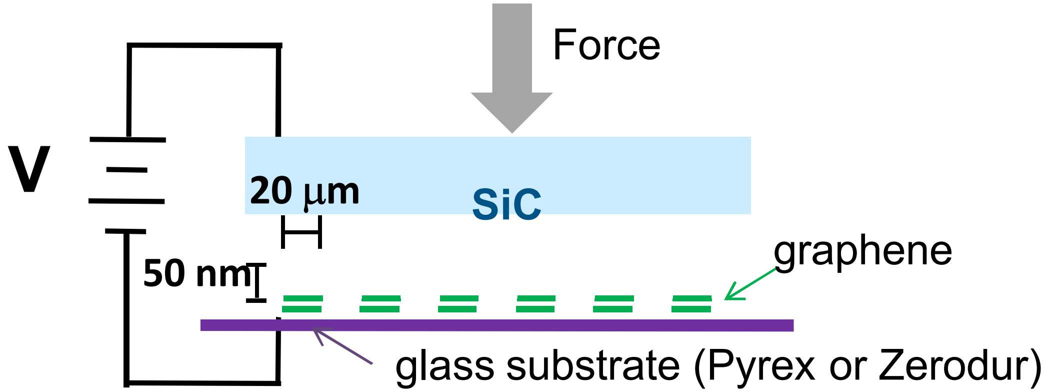
Graphene transfer procedure

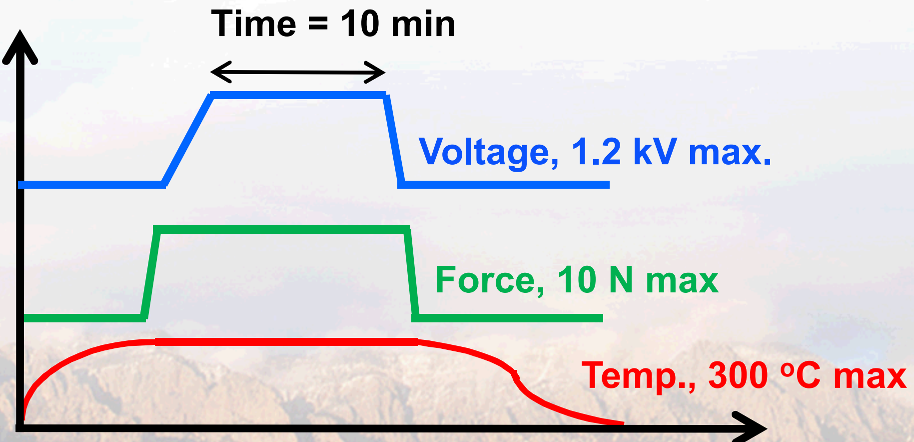
As-grown C-face graphene



Patterned graphene







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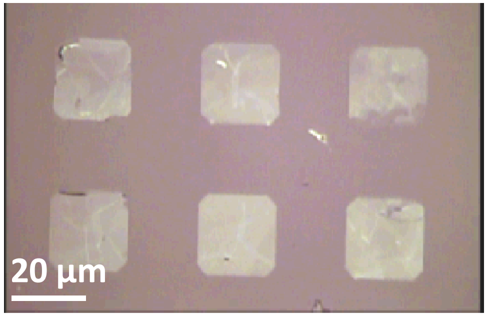
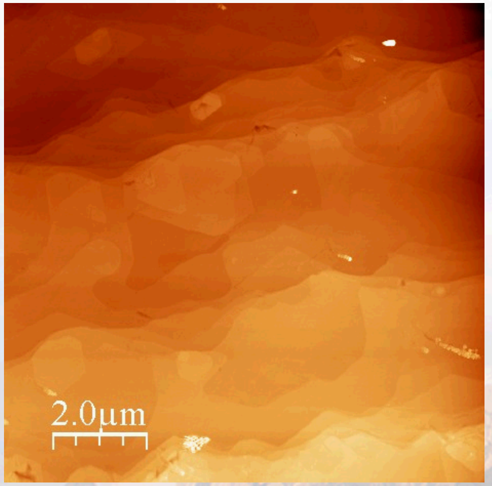
How to judge graphene transfer

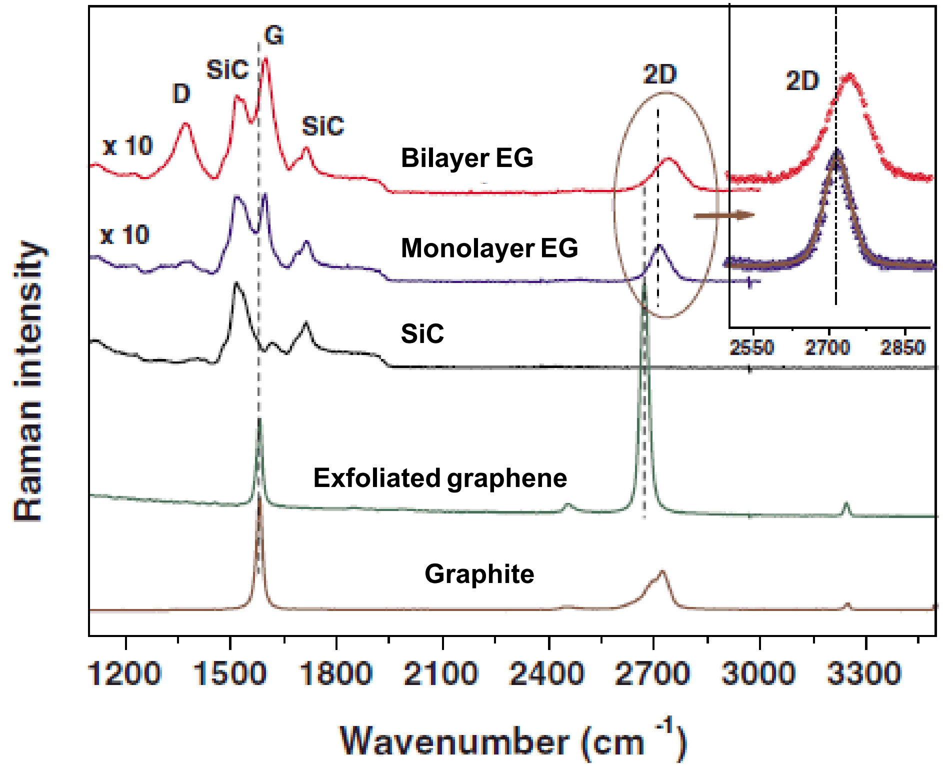
Optical microscopy



Atomic force microscopy



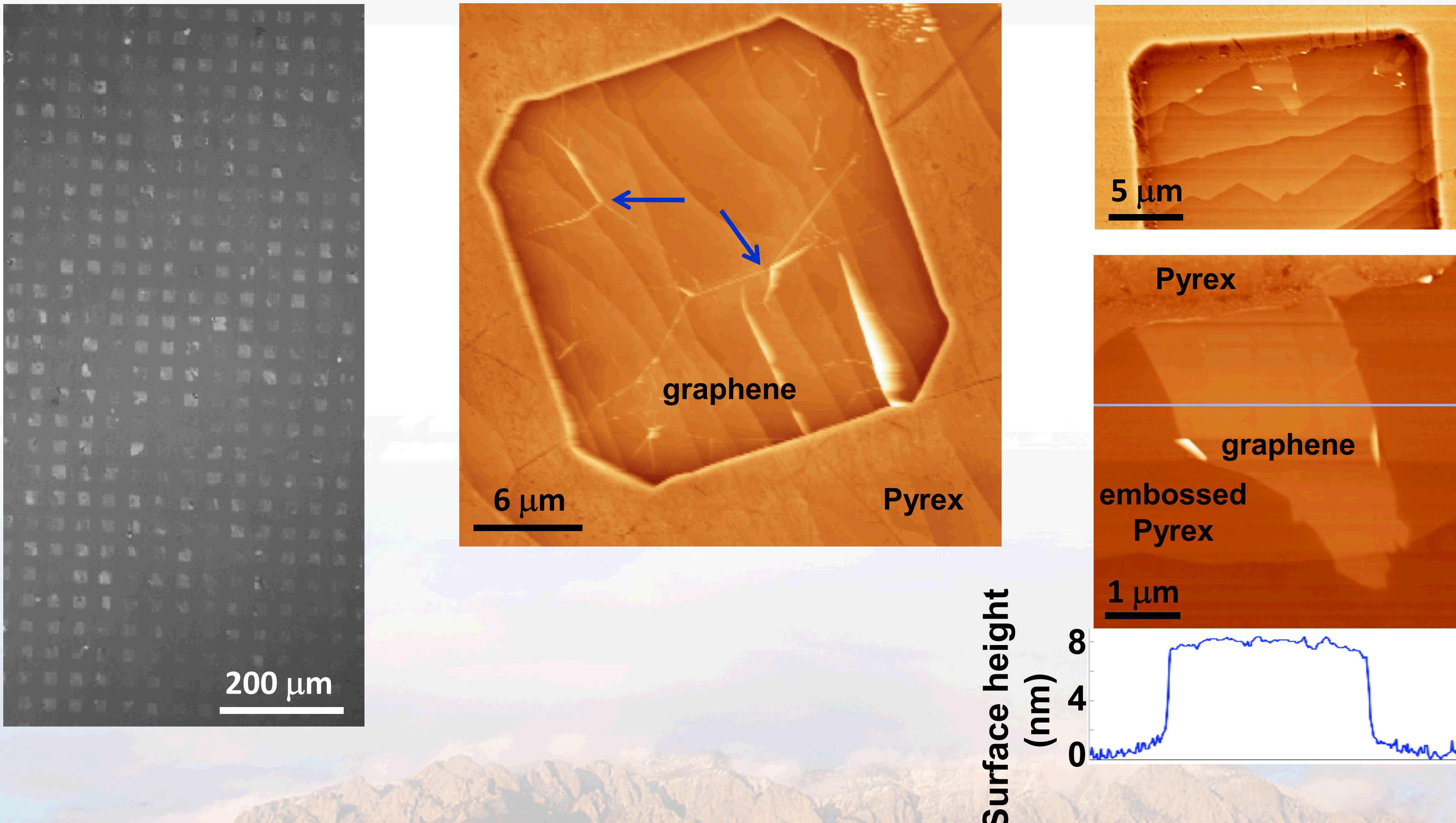
Raman spectroscopy



D-band: ~1350 cm^{-1}
G-band: ~1580 cm^{-1}
2D-band: ~2700 cm^{-1}

Z. H. Ni *et al., Phys Rev B* **77**, 115416 (2008)

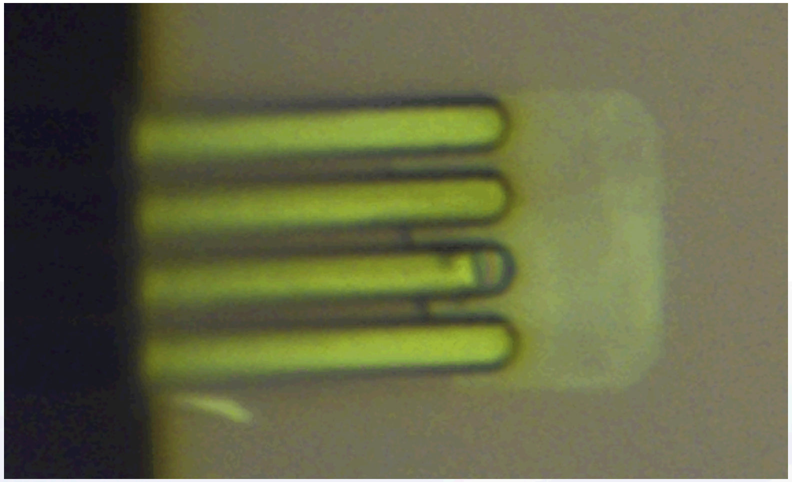
C-face graphene squares transferred to Pyrex



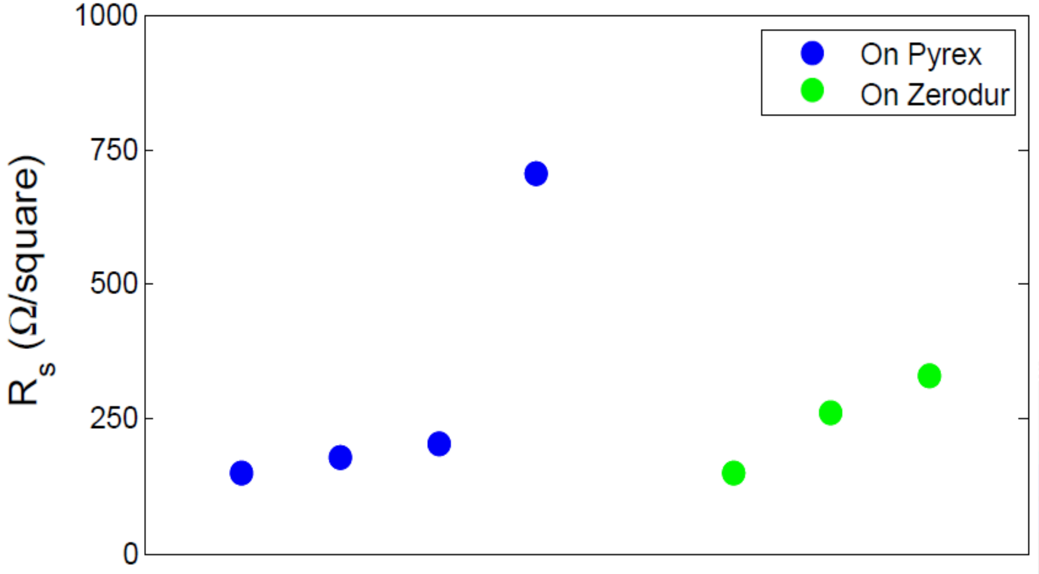
Electronic characterization of transferred graphene from SiC (C-face)

$$R_s = \frac{\pi}{\ln 2}\frac{V}{I}$$

Graphene transferred from the C-face has R_s as low as 150 Ω/□.



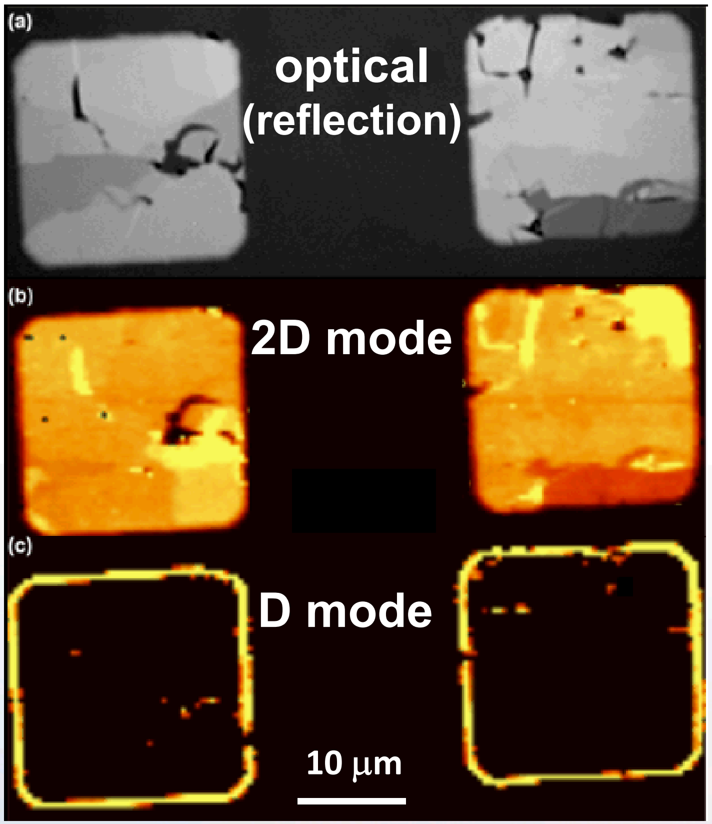
Micro four probe on 10 x 10 μm^2 square of transferred graphene

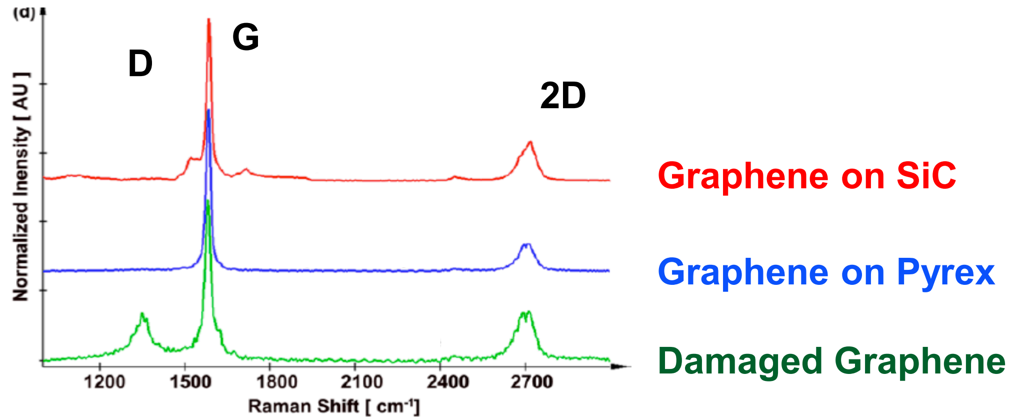


L. Biedermann, T. Beechem, A. Ross, T. Ohta, and S. Howell, submitted to *New J. of Physics.*

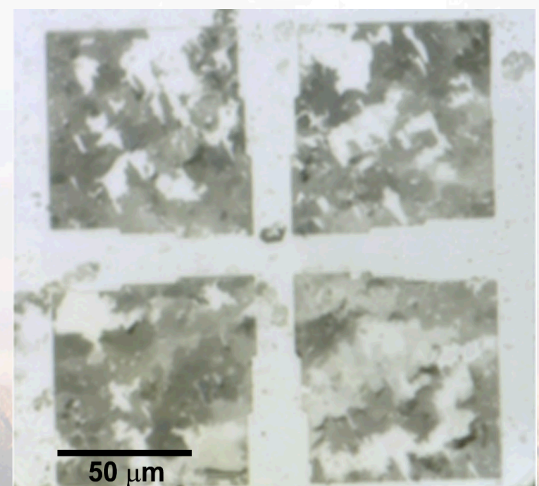
Raman analysis confirms graphene transfer

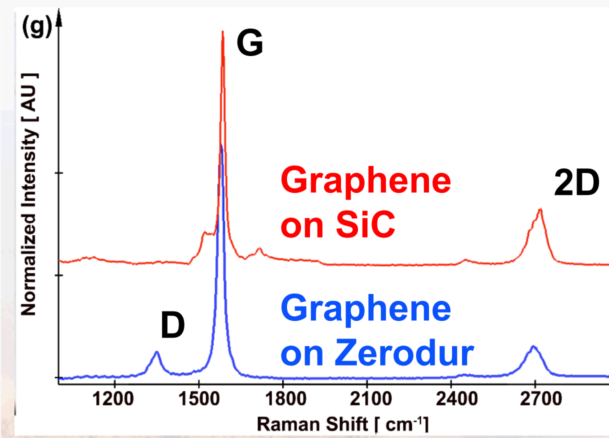
Graphene on Pyrex





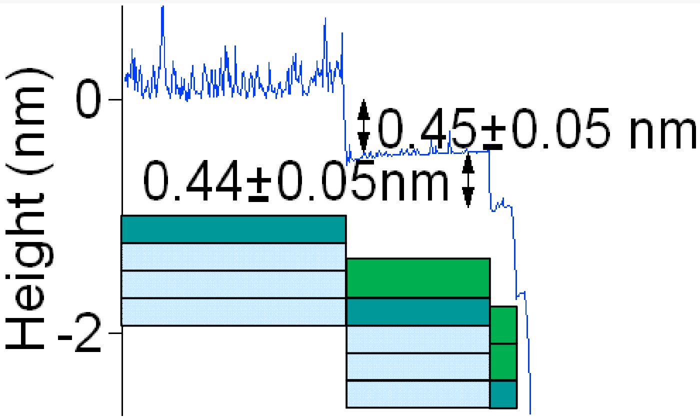
Graphene on Zerodur

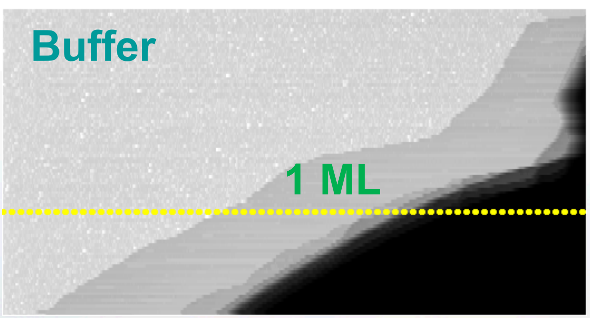




L. Biedermann, T. Beechem, A. Ross, T. Ohta, and S. Howell, submitted to *New J. of Physics.*

About-face: Si-face epitaxial graphene

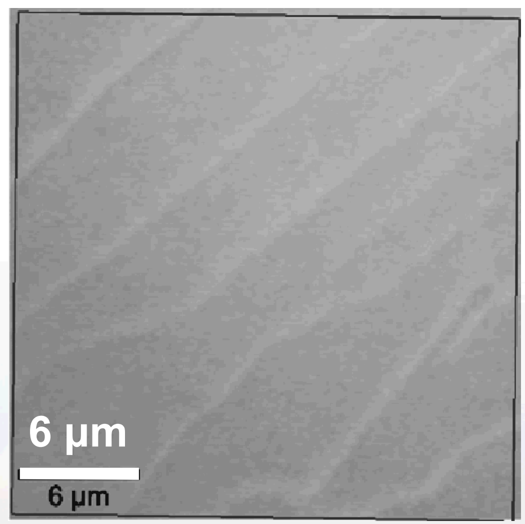




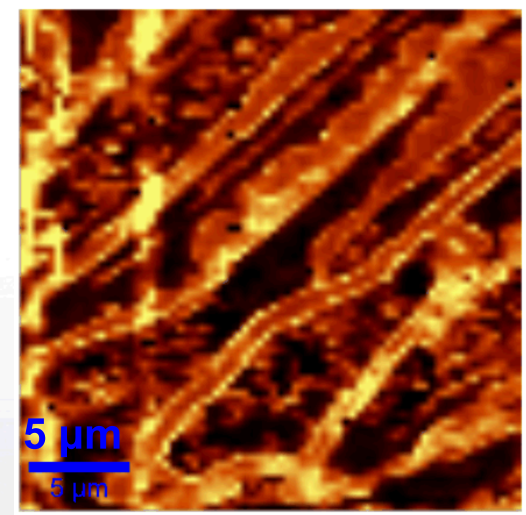
T. Ohta *et al. Phys Rev B* **81,** 121411(R) (2010).

Sample 090728 #27
60 % 1 ML, 30 % 2 ML, 10 % ≥ 3 ML

Optical

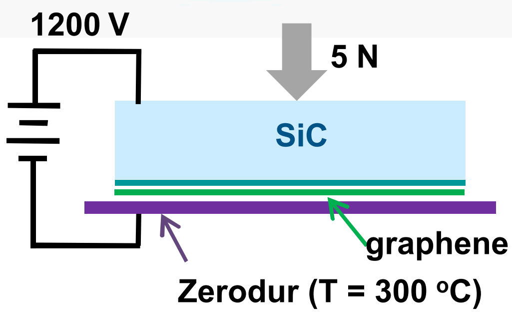


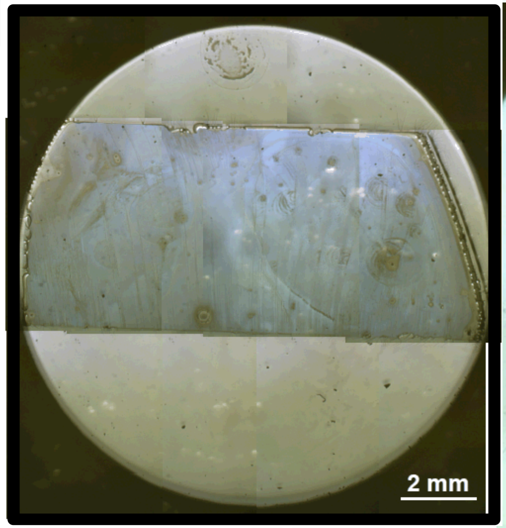
2D FWHM

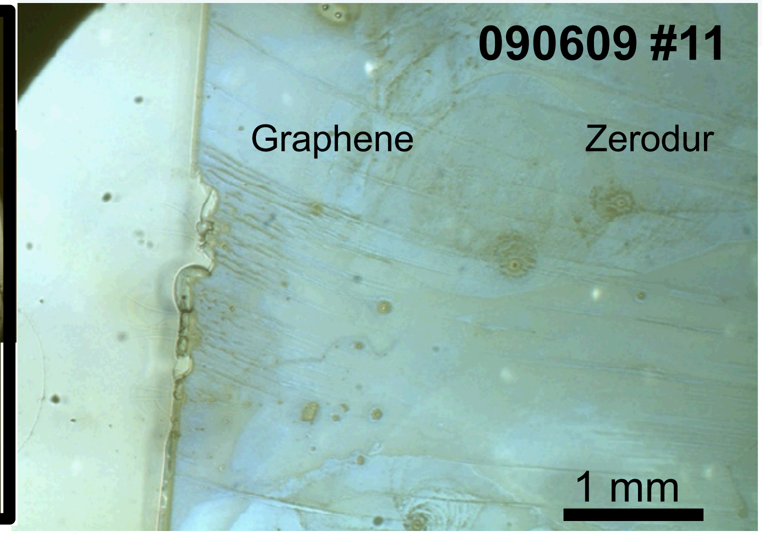


Mobility at 4 K: μ = 14000 cm²/Vs
Wei Pan, Sandia

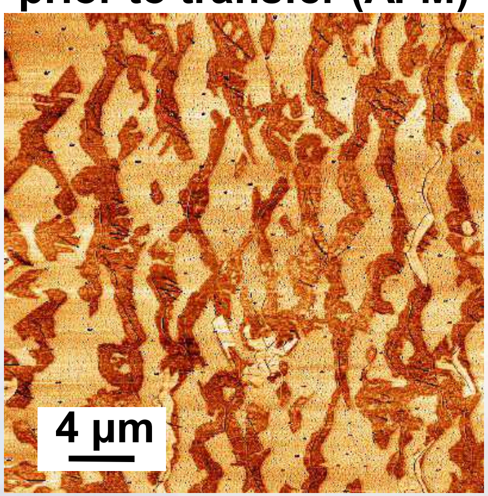
Chip-scale transfer of Si-face graphene to Zerodur







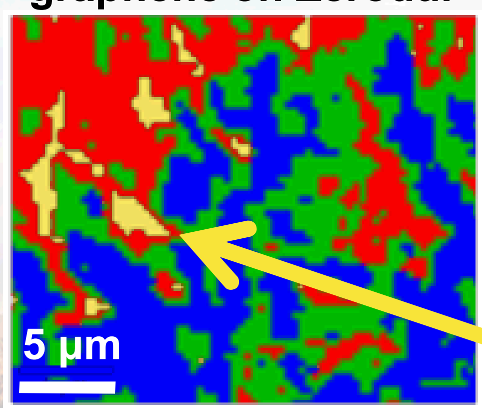
Graphene on SiC, prior to transfer (AFM)

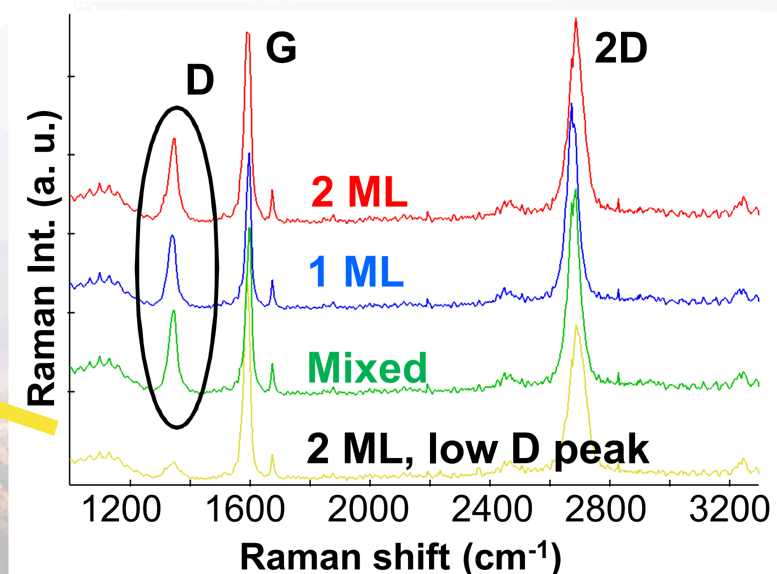


Bright phase = 1 ML

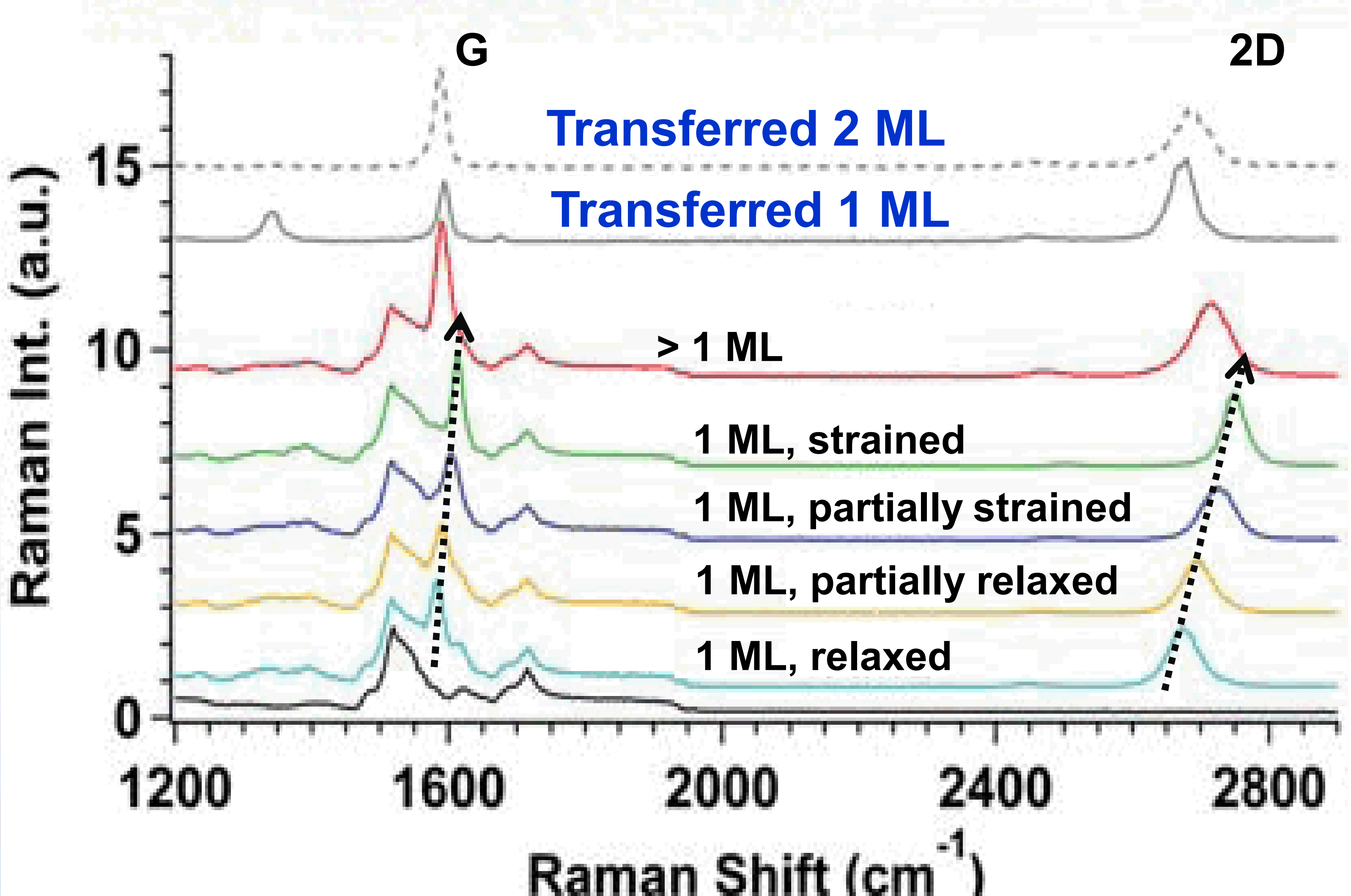
Dark phase = 2 ML

Cluster analysis of graphene on Zerodur





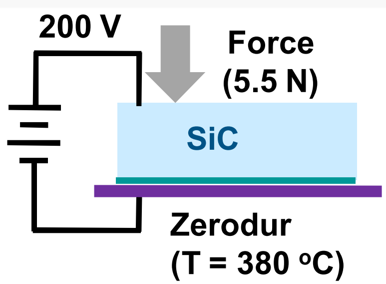
Transfer relaxes strain inherent in epitaxial graphene

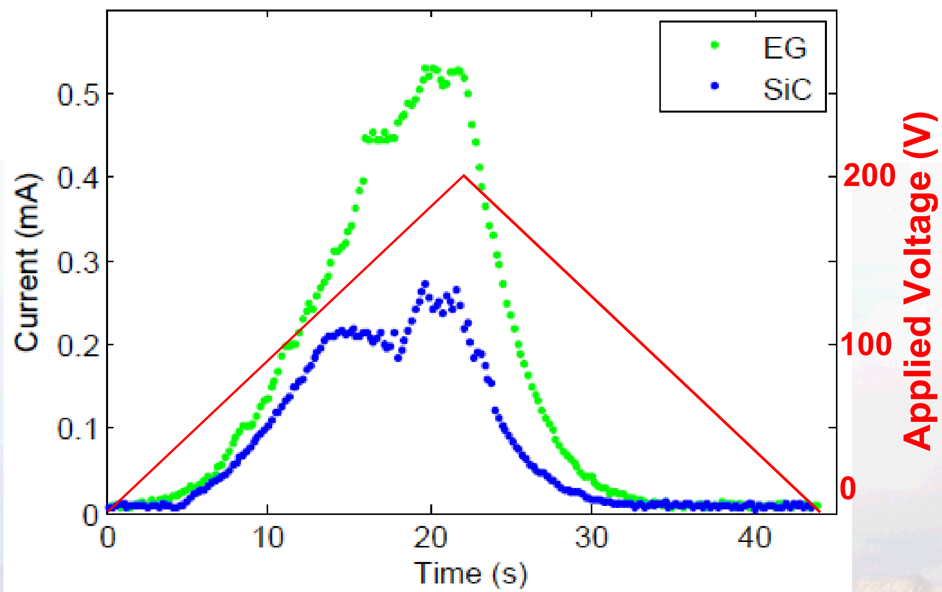


D. Schmidt *et al.,* submitted to *Nano Letters* (2010).

Electrochemical analysis suggests graphene is bonding to the Zerodur

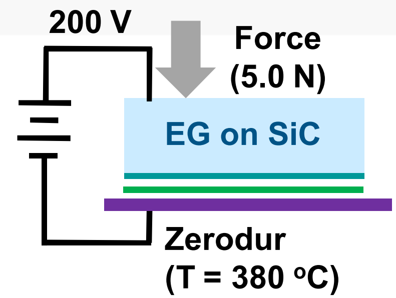
Anodic bonding of SiC to Zerodur

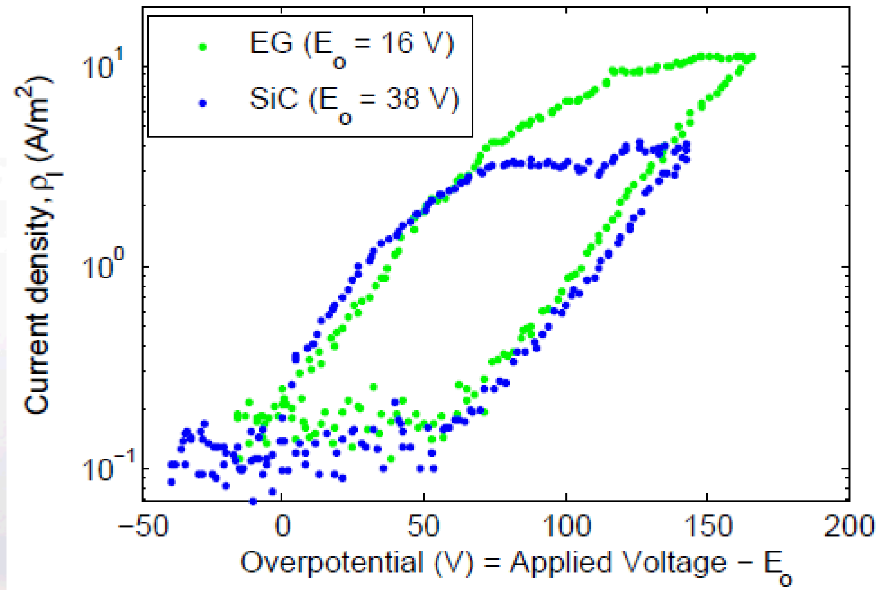




- Zerodur cannot be removed without breaking SiC

Transfer of EG to Zerodur

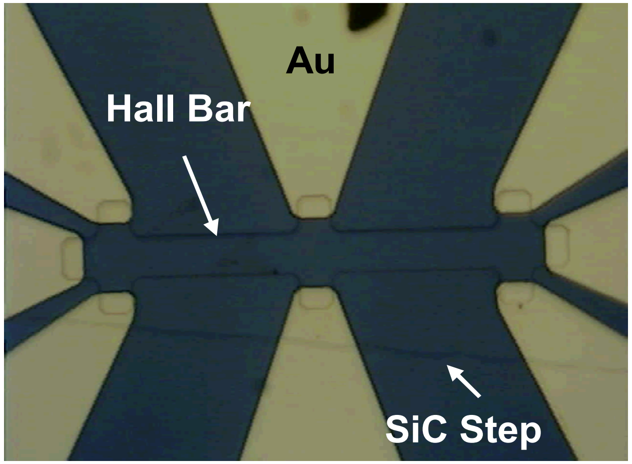




- Zerodur releases from graphene/Zerodur with little force

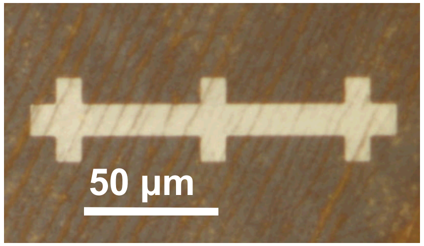
Comparison of transferred Si-face graphene to EG

EG Hall bar on SIC(0001)



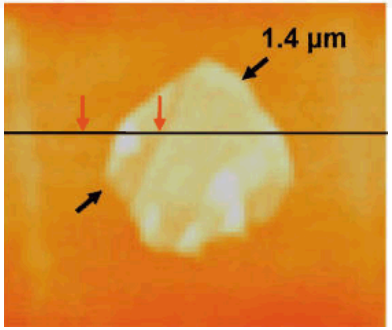
- Epi-graphene electron mobility: 14,000 cm^2/Vs
- Electron density: 6 x 1011 cm^2
- Graphene sheet resistance: ~1600 Ω/sq (average from 12 devices)

Hall bars structures, Si-face graphene on Zerodur



- Poor large scale transfer (most Hall bars are open circuit across their length)
- Measurements across small distances (< 10 µm) do show some electrical activity (~ 5 kΩ - 5 M Ω)
- The transferred material appears to be isolate ribbons of graphene
  - Breaks could be caused by SiC steps

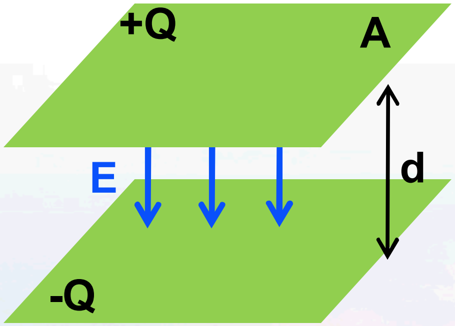
Ongoing work: Electrostatic rather than electrochemical transfer



Pre-patterned HOPG to SiO_2
8.5 V
$\mu \sim 1050\ cm^2/Vs$

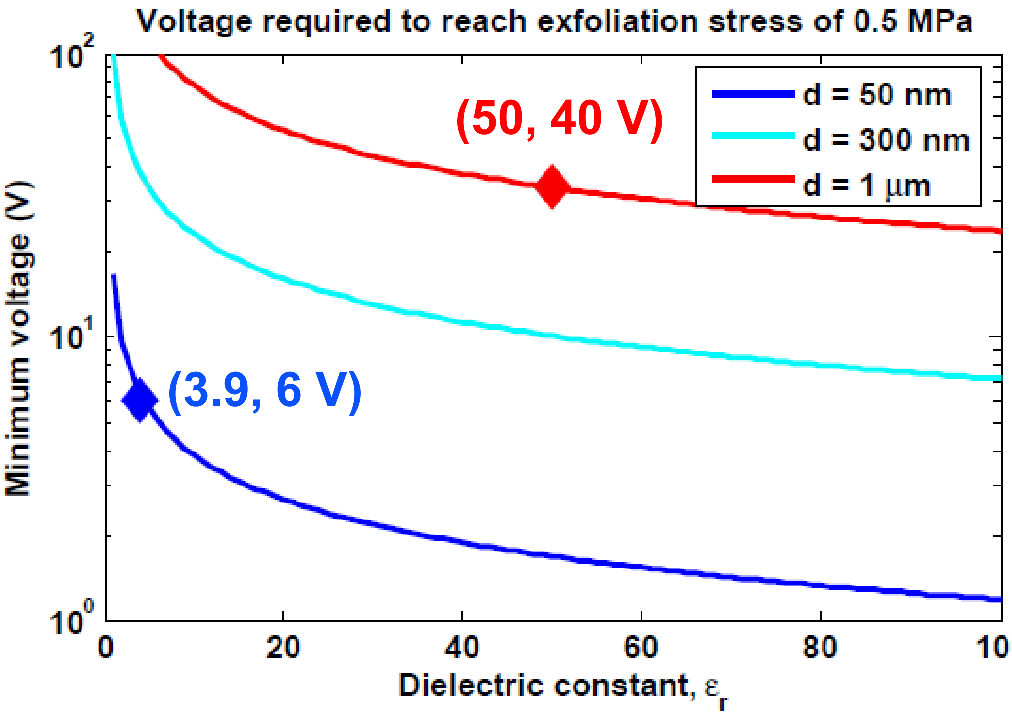
X. Liang *et al.*, *Nano Lett.* **9**, 467-72 (2009).

The exfoliation strength of graphite is ~ 0.4 MPa.



$$\vec{F}_{cap} = Q\vec{E}$$

$$P_{cap} = \frac{\vec{F}_{cap}}{A} = \frac{\varepsilon_o \varepsilon_r V^2}{d^2}$$



Conclusions

- **Demonstrated transfer of graphene to Pyrex and Zerodur from the C-face of SiC**
- **Graphene transferred from C-face was characterized with Raman and mico four probe**
  - The average resistance was found to be XXX Ω/Sq
- **Attempted transfer of graphene monolayer from Si-face of SiC**
- **Characterized with Raman Spectroscopy**
  - Detected presence of defects
  - Showed relaxation in strain
- **Electrical charterization determined that the transferred films were not continuous over large length scales (transfer of ribbons)**
- **Current monitor during transfer indicated possible chemical bonding to Zerodur**

Graphene collaborators

Laura Biedermann, Transfer

Thomas Beechem, Raman spectroscopy

Taisuke Ohta, epitaxial graphene growth

Anthony (TJ) Ross, lithography and etching

Jeff Stevens, etching

Graham Yelton, electrochemistry advice

Backup Slides

Explored transfer parameter space; transfer still damages graphene

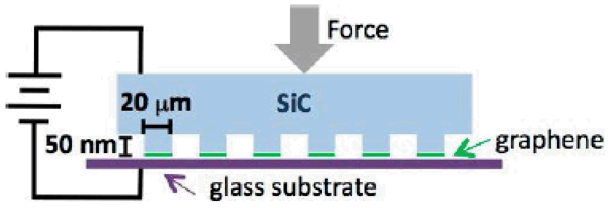
	Range	090609 #11	081209 #30	090609 #33	090728 #06	090401 #03	0907278 #18
Voltage (kV)	0.2 – 1.2	1200	530	525	525	550	550
Force (N)	2 – 10	5.0	4.0	4.2	3.9	8.9	7.7
Time (min)	1 – 10	10	4	3.5	1	4	5
Temp (°C)	300 – 390	300	370	370	370	390	370
% 1 ML	45 – 85	50	45	80	80	85	75
I(D) / I(G)		1.0	1.3	1.0	0.7	1.4	1.2

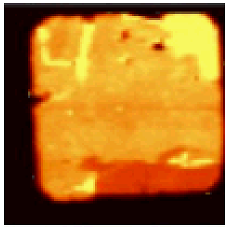
Metric for graphene quality:

$$\frac{I(D)}{I(G)} = \frac{\text{D-peak intensity}}{\text{G-peak intensity}}$$

Graphene transfer

- Scanning tunneling microscopy and atomic force microscopy
- **Graphene transfer to insulating substrates**
- Laser Doppler Vibrometry
- Casimir force measurements

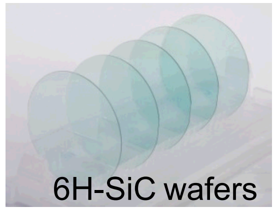


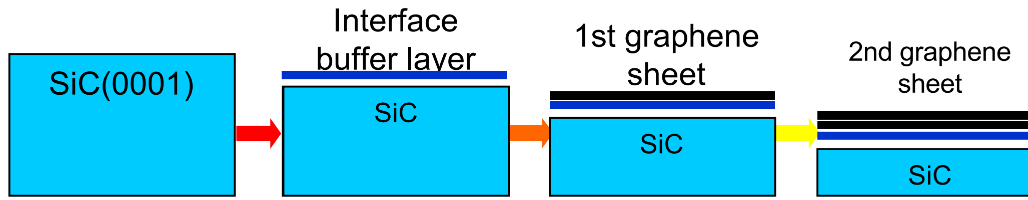


- **LDRD 11-0876: Enabling Graphene Nanoelectronics**
- **PI: Stephen W. Howell, PM: Carlos Gutierrez**
- **Core team:** Thomas Beechem, Tom Friedmann, Tim Lambert, Kevin Leung, Kevin McCarty, Taisuke Ohta, Wei Pan, Anthony Ross, Jeff Stevens, Cody Washburn, and David Wheeler
- **Publications submitted:**
  - L. Biedermann, T. Beechem, A. Ross, T. Ohta, and S. Howell, "Electrostatic transfer of patterned epitaxial graphene from SiC(000-1) to glass," submitted to *New J. of Physics.*
  - D. Schmidt, T. Ohta, L. Biedermann, S. Howell, and G. Kellogg, "Inhomogeneous strain fields in epitaxial graphene," submitted to *PNAS.*

Understanding Graphene Growth on SiC

- **Graphitization of SiC:**
  - Sublimation of Si at high temperature (>1200 ^{o}C) leaves graphene layer at SiC surface





•Argon-assisted graphene formation yields large (~100 μm^2) graphene domains

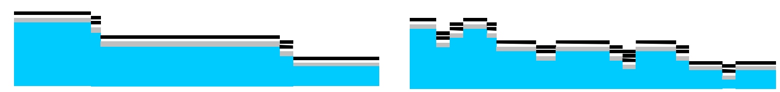
•Samples prepared using Ar atmosphere at atmospheric pressure and high temp

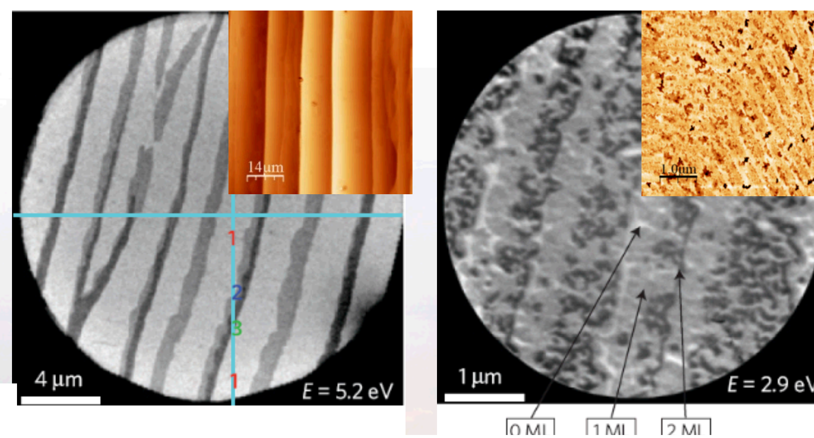
•Using this method we have exquisite control of :

•Domain size

•Percentage coverage of mono/bilayers

Atmospheric pressure Ar high temp. processing | **Ultrahigh vacuum mid temp. processing**

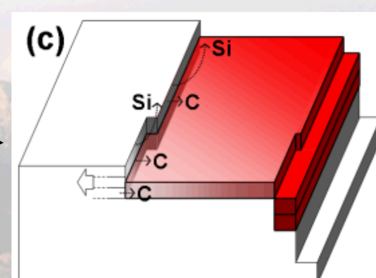




•Understanding the growth mechanism of graphene on SiC

•Growth morphology strongly depends on the step structure

Step-flow growth
•Key for high large-area growth
•Real-time growth observations using LEEM



K. V. Emtsev et al., Nature Mater. 8, 203 (2009).
C. Virojanadara et al., Phys. Rev. B 78, 245403 (2008)

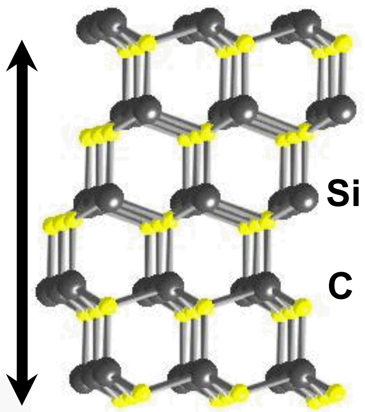
T. Ohta, N. C. Bartelt, S. Nie, K. Thürmer, G. L. Kellogg, PRB 81, 121411(R)(2010)

Epitaxial graphene growth on SiC

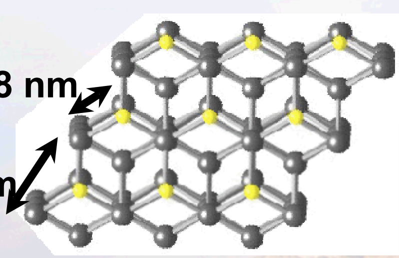
4H - SiC

Si face (0001)

1.0 nm



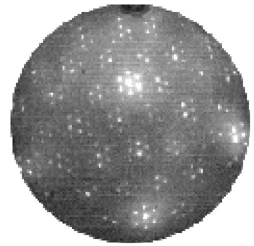
C face (000-1)



1050 °C

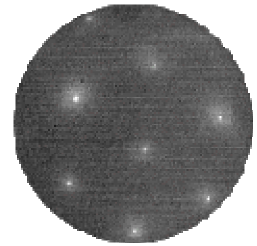
SiC

1150 °C



Buffer layer

1400 °C

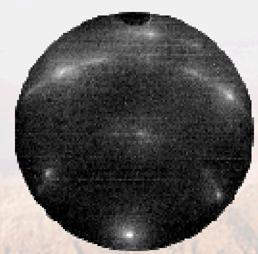


Single-crystal graphite

- 1 to few ML graphene
- Buffer layer between graphene and SiC

Increasing temp.

1200 °C



polycrystalline graphite

- Many-layer graphene common
- Disordered stacking

I. Forbeaux *et al.*,*Surf. Sci.* **162-163**, 406 (2000)