SAND2011-1903C

Characterization and comparison of devices fabricated from epitaxial graphene on SiC and electrostatically transferred graphene

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Sandia National Laboratories

GOMAC Tech 2011

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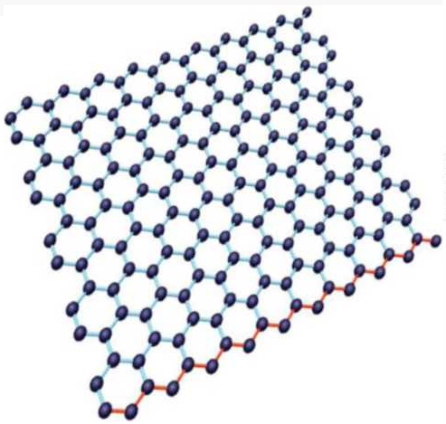
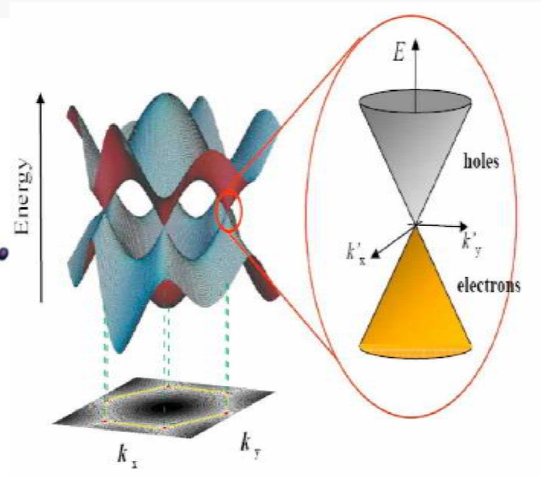
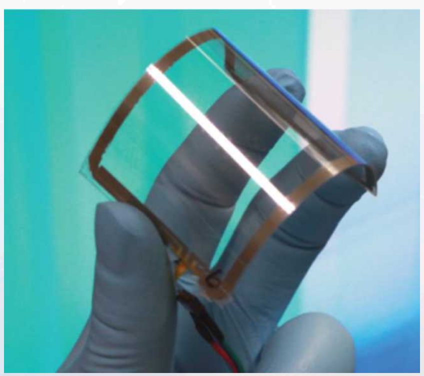
Why graphene?





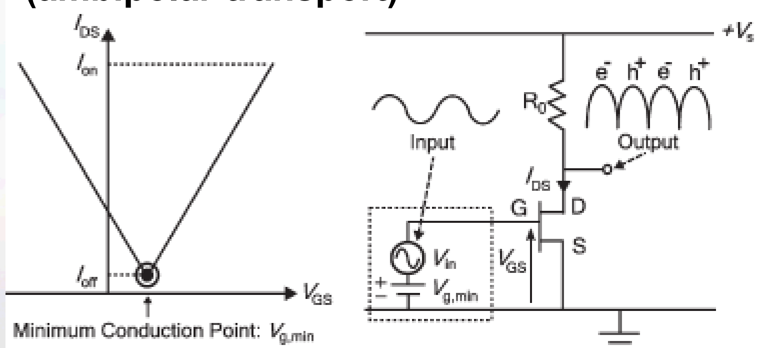
Mobility, μ, up to 250,000 cm^2/Vs
(suspended exfoliated graphene)
Ambipolar, zero-bandgap
Current densities up to 5 x 10^8 A/cm^2
Elastic modulus ~ 1 TPa

Transparent electrodes



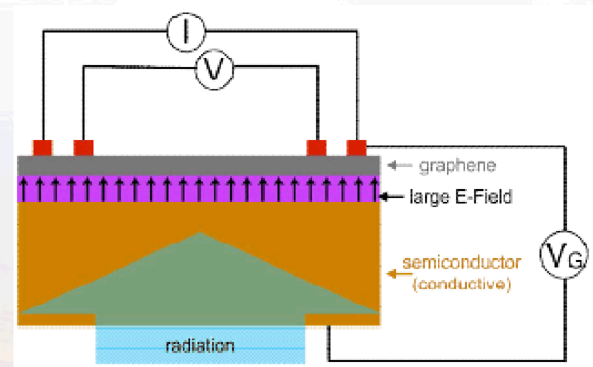
S. Bae *et al, Nature Nano.*, **5**, 574-578 (2010)

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).

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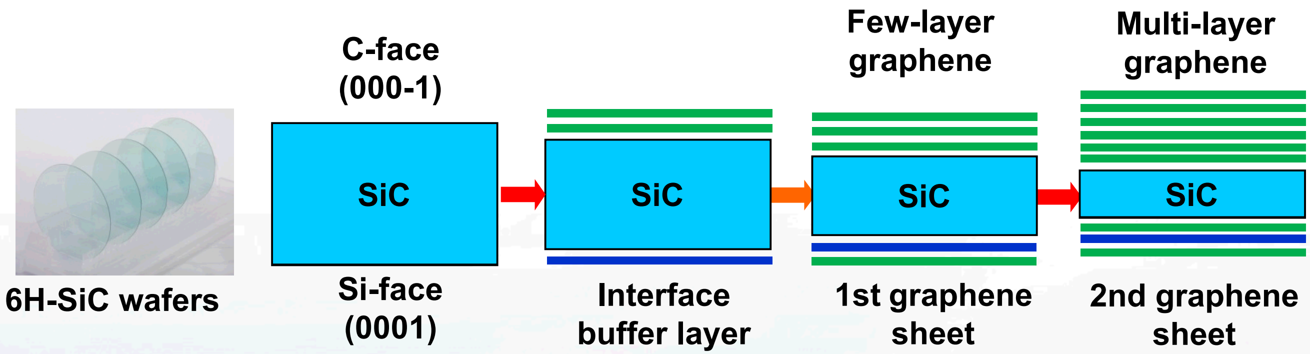
Motivation for transferring epitaxial graphene (EG) from SiC

- **Transfer of CVD grown graphene has been demostrated**
  - CVD typically has lower mobilities than EG on SiC
- **Limitations of EG on SiC**
  - Lack of a back gate
  - Difficult to create suspended graphene
  - EG is inherently doped by underlying SiC (electronic properties are highly substrate dependent)
- **Why use EG on SiC?**
  - EG grown on SiC has high carrier mobility
  - Current synthesis approaches allow for tight control of graphene thickness (wafer-scale monolayer and bilayer coverage)
- **Development of a scalable transfer technology <u>that preserves EG's remarkable electronic properties</u> could enable:**
  - EG integration with current CMOS processes
  - Integration with optical devices (eg: metamaterials, laser pulse formers)

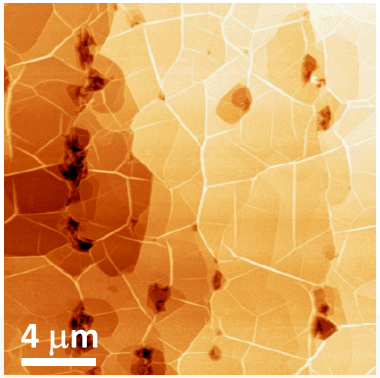
Ar-mediated synthesis yields high quality epitaxial graphene

SiC graphitization conditions:

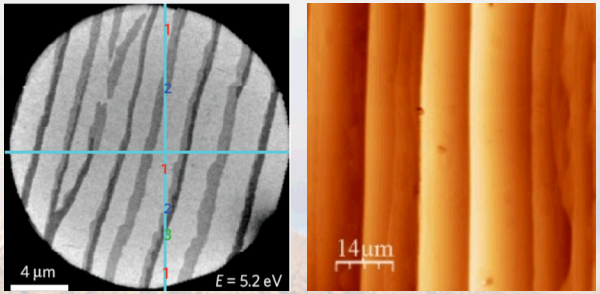
- High temperature (>1200 °C)
- Argon atmosphere at atmospheric pressure



C-face (000-1)



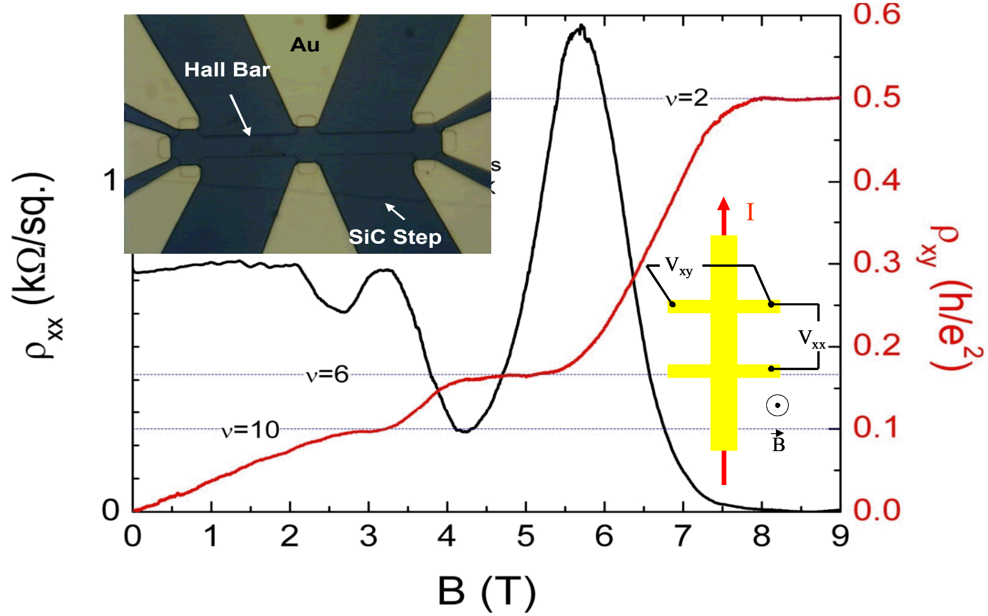
Si-face (0001)



K. V. Emtsev *et al., Nature Mater*. **8**, 203 (2009)..
C. Virojanadara *et al., Phys. Rev. B* **78**, 245403 (2008).
T. Ohta *et al., Phys Rev B* **81** 121411(R) (2010).
Samples provided by T. Ohta

Electronic characterization of EG grown on SiC (0001)

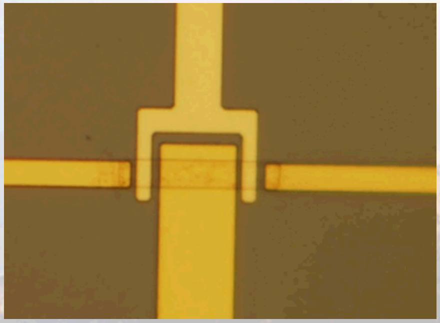
Low Temp Transport Measurements (4 K)

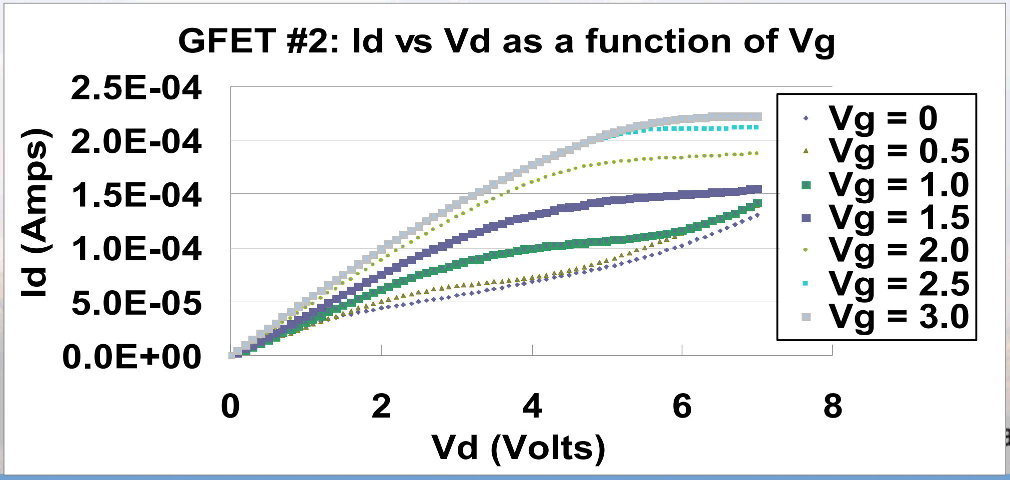


- EG electron mobility: 14,000 cm^2/Vs
  - **Record mobility** (at time of measurement)
- Electron density: 6×10^{11} cm^{-2}
- Graphene sheet resistance:
  - **~1600 Ω/sq** (average of 12 devices)
  - Indicates high uniformity
- Observed IQHE on 3 devices on the same chip

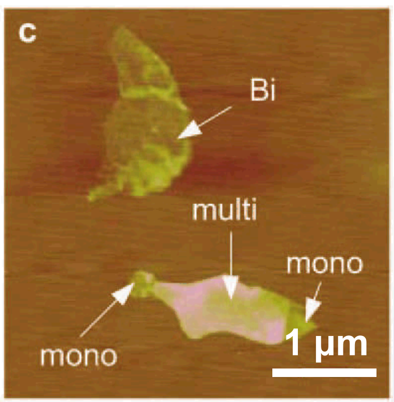
Pan, Howell, Ross, Ohta and Friedmann
Appl. Phys. Lett. 97, 252101 (2010)

GFET Development





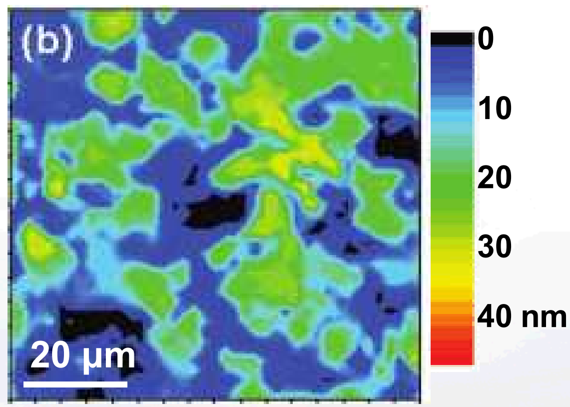
Methods to transfer EG to SiO_2



Si-face EG
Small flakes

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

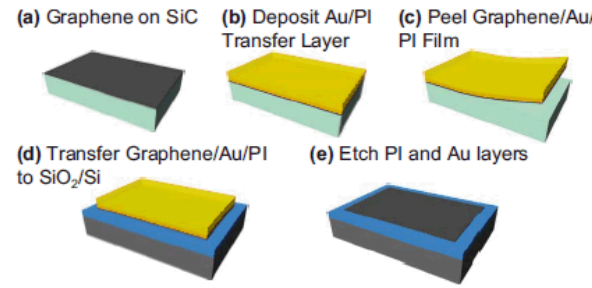
Thermal release tape
+ 5 N/mm² pressure

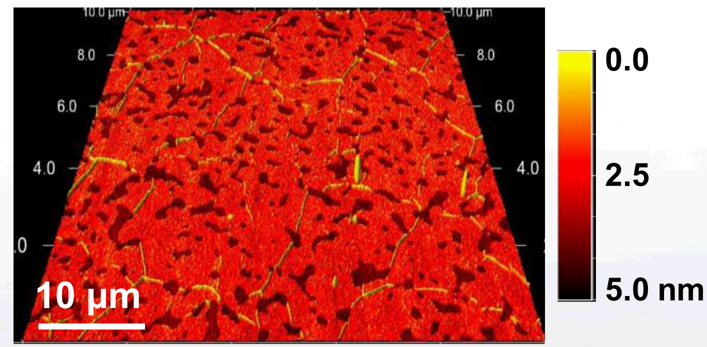


C-face EG
Very thick graphene
μ ~ 1350 cm²/Vs

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

Gold/polyimide film handle





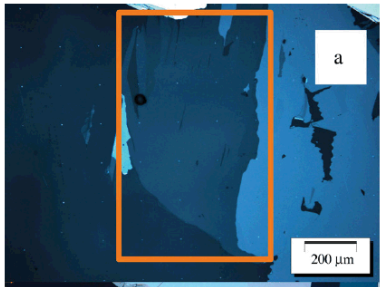
Si-face EG
Damaged graphene
μ ~ 100 cm²/Vs

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

Voltage-driven exfoliation of graphite

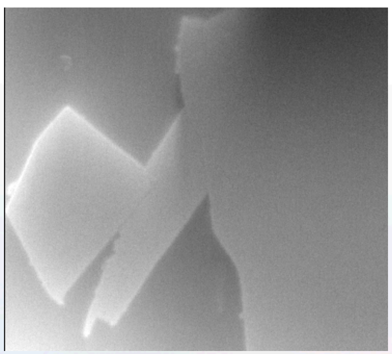
Stress to exfoliate graphene from bulk graphite: P = 0.4 MPa

T. Kuzumaki *et al., APL*, 79, 4580-2 (2001).



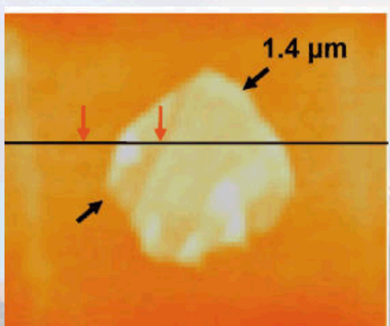
Graphite to Pyrex
1.2 – 1.7 kV
$\mu \sim 10^4$ cm^2/Vs

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



HOPG to SiO_2
3-30 kV

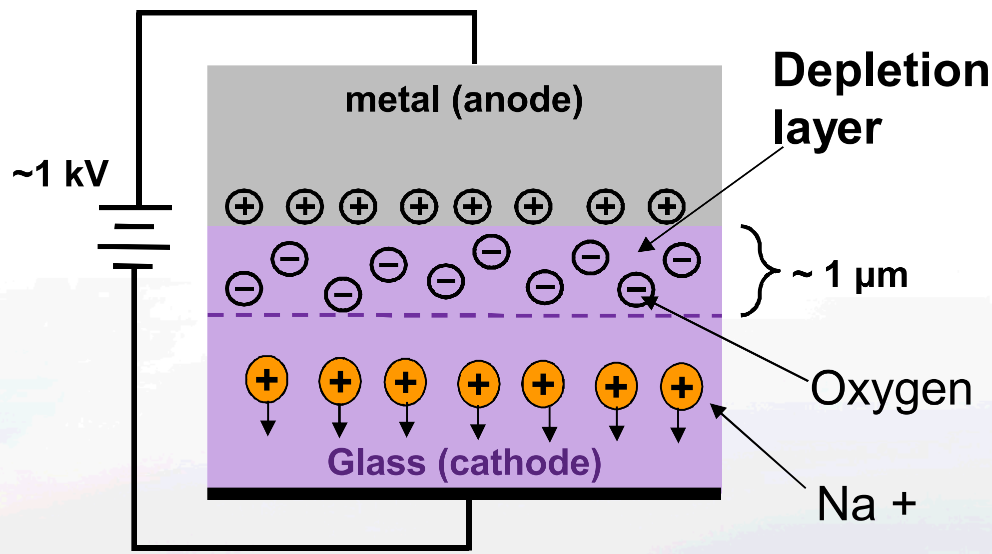
A. Sidorov *et al., Nanotechnology* **18**, 135301 (2007).



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

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

Shukla's method is derived from anodic bonding.

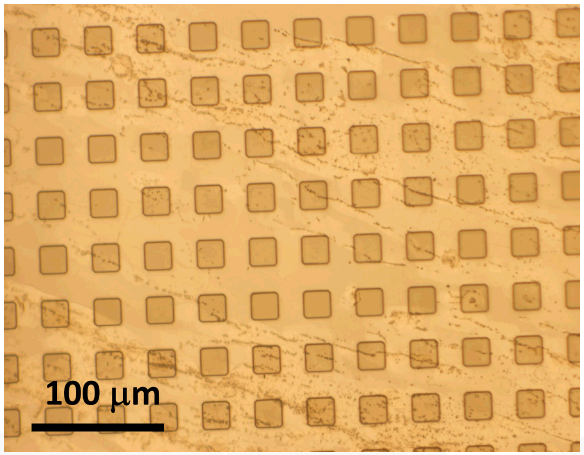


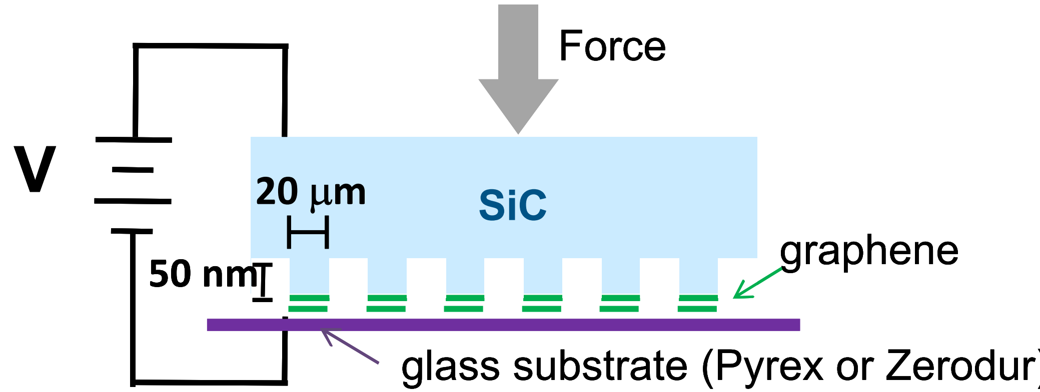
$$E_{depletion} \approx 300\ \mathrm{MV/m}$$

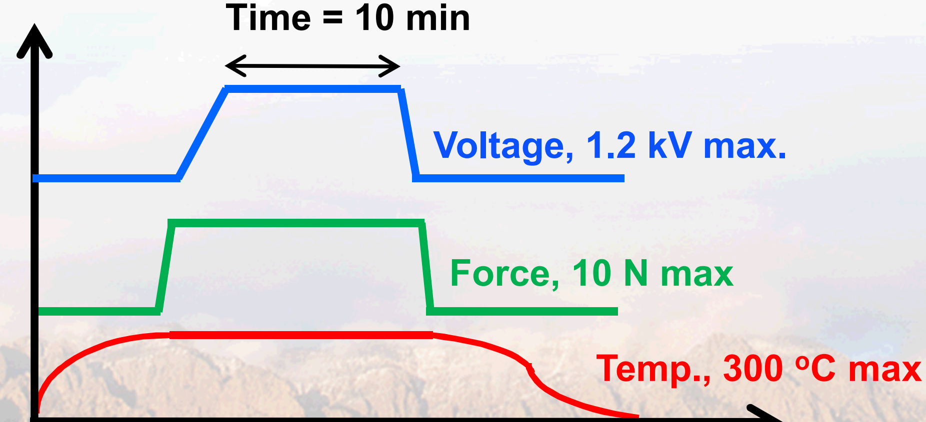
Assuming parallel plates: $P = \frac{1}{2}\varepsilon_o \varepsilon_r E^2 \approx 2\ \mathrm{MPa}$

Graphene transfer procedure

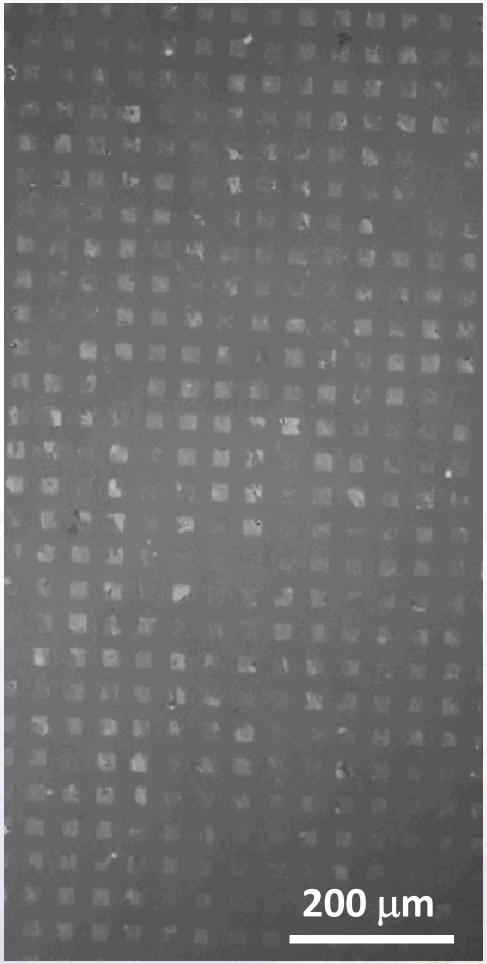
Patterned epitaxial graphene

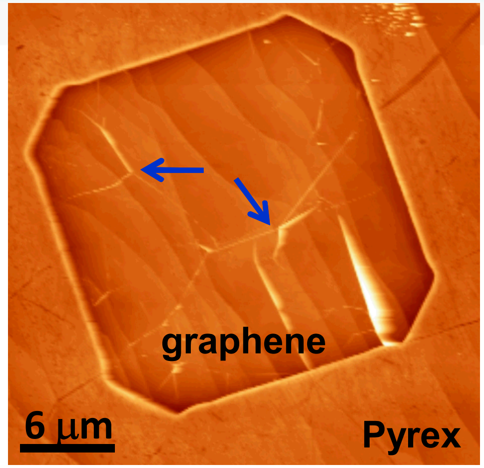


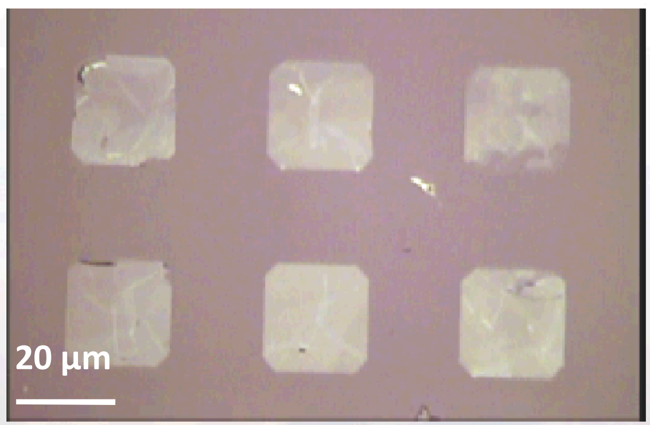


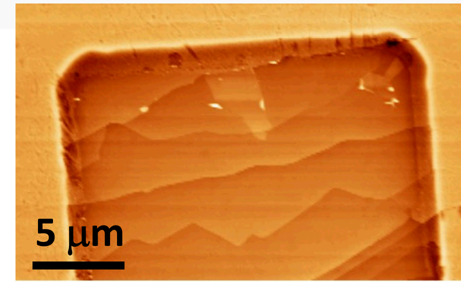


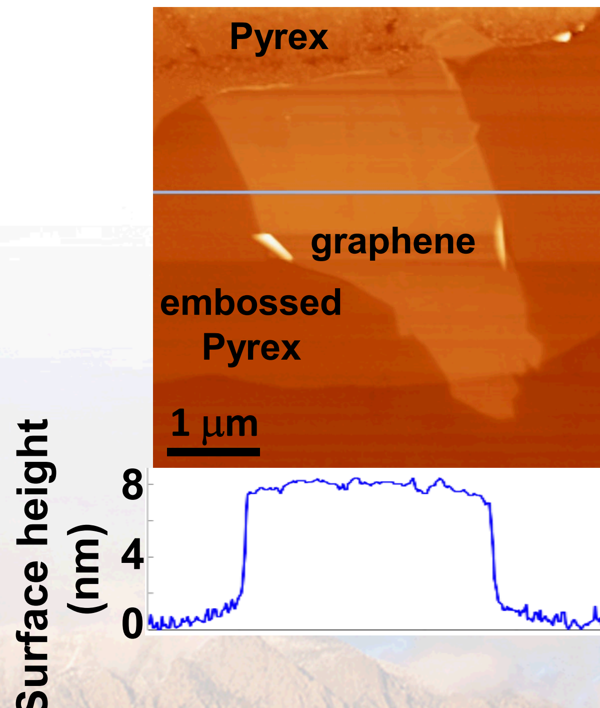
C-face graphene squares transferred to Pyrex







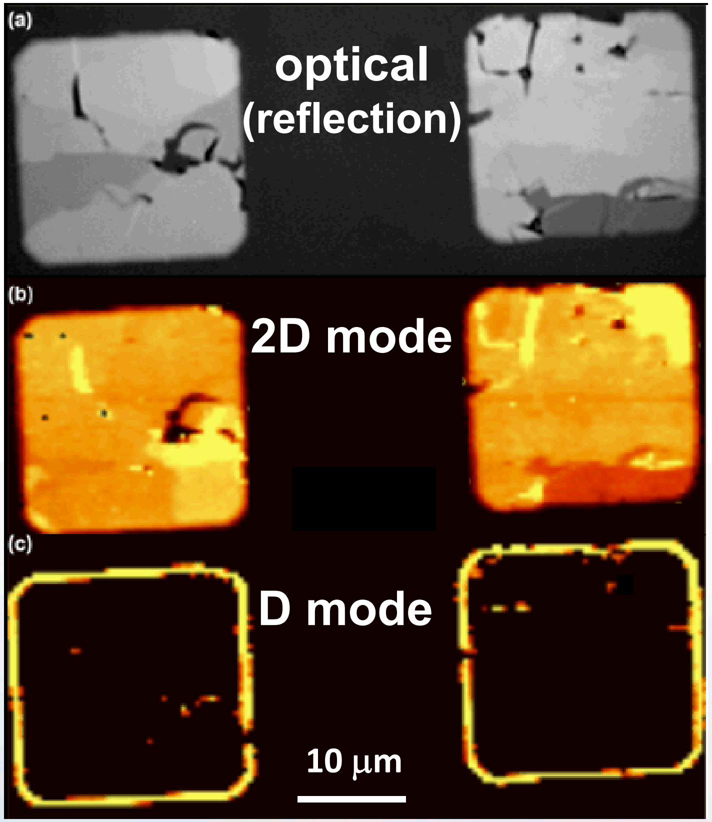


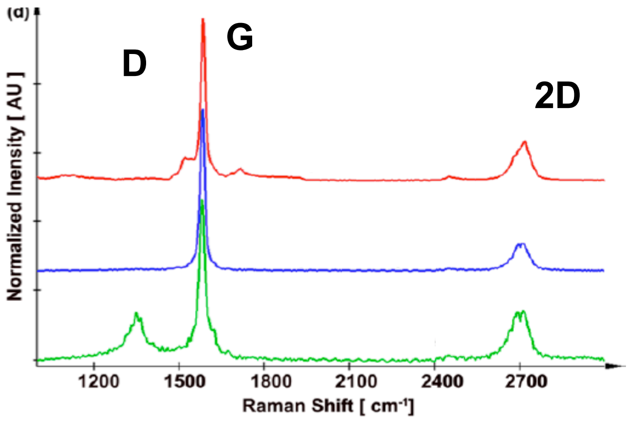


63% ± 7% of graphene was successfully transferred (determined by flood analysis of optical image)

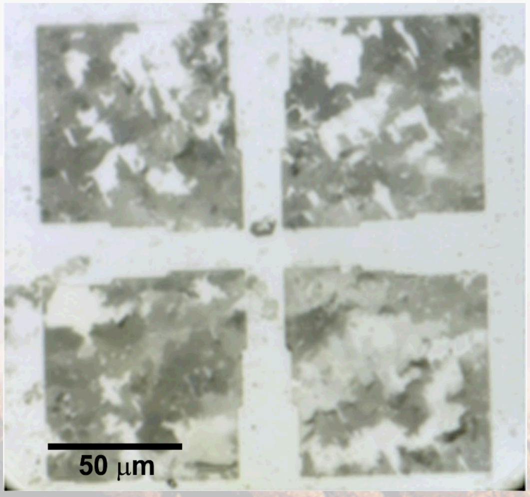
Raman analysis confirms graphene transfer

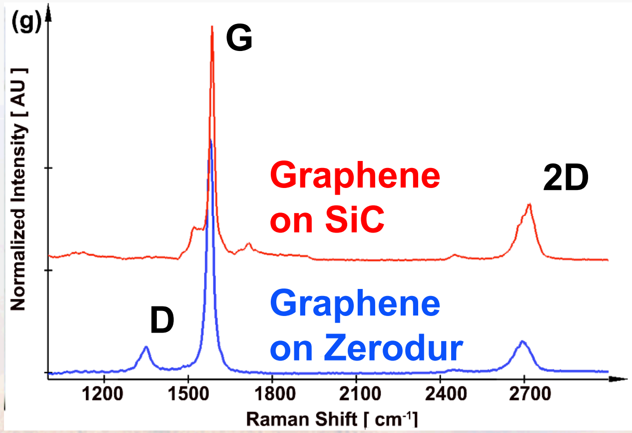
Graphene on Pyrex





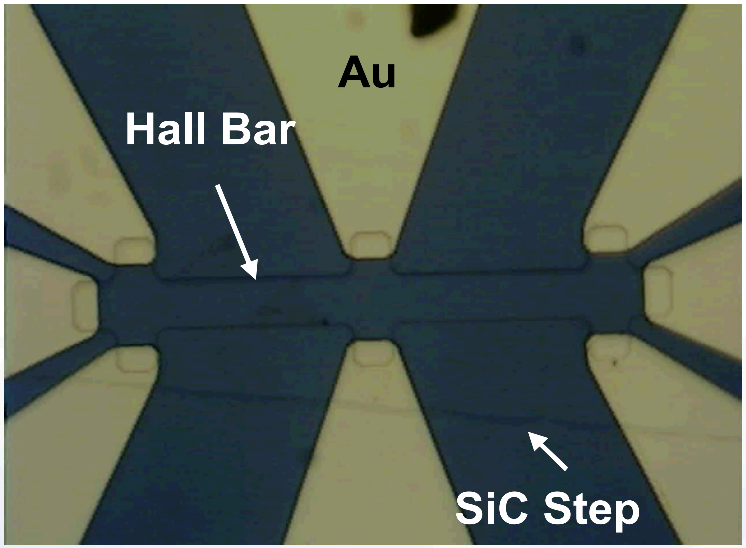
Graphene on Zerodur





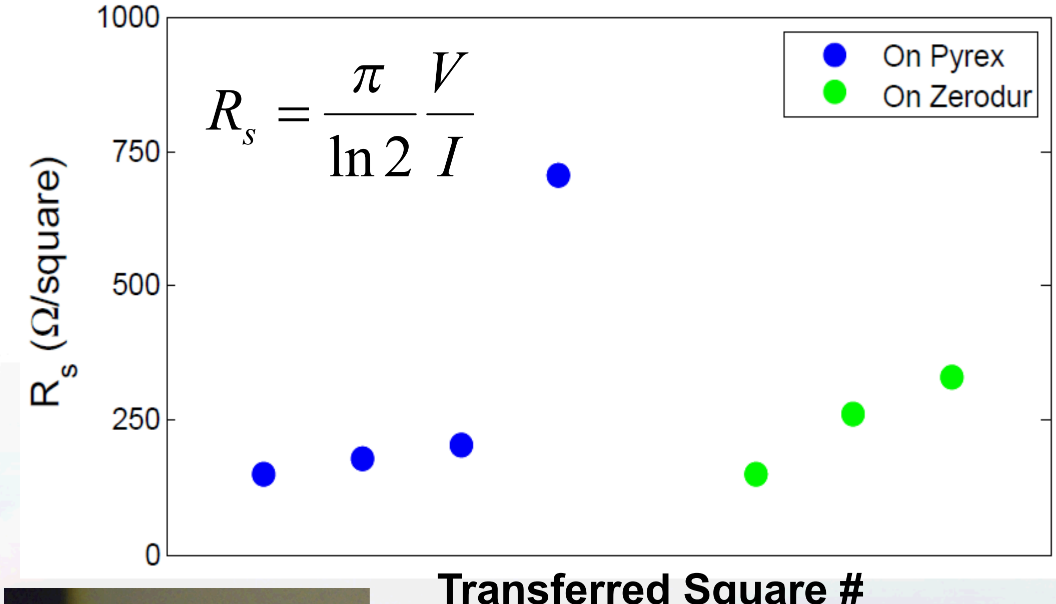
Electronic characterization of C-face transferred graphene

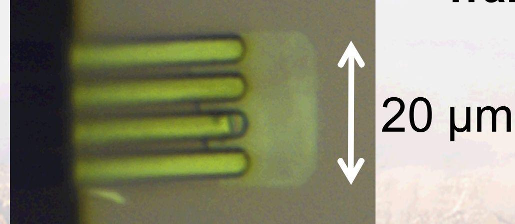
At ~10 K, R_s = 180 ± 70 Ω/sq for epitaxial graphene on SiC(000-1)



Sizes: 400 x 50 μm^2 down to 50 x 6 μm^2

At RT, R_s is as low as 150 Ω/sq for transferred graphene

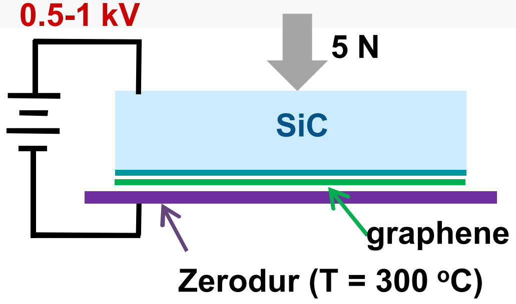




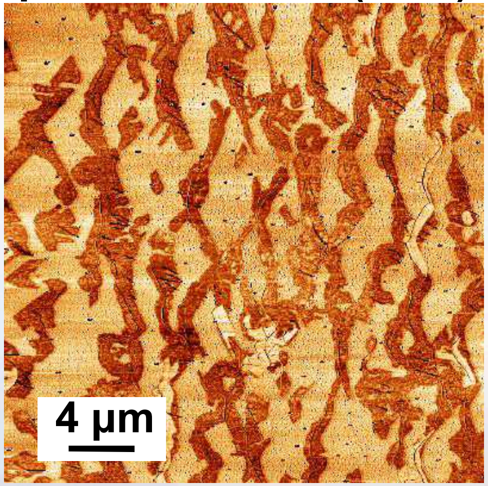
- Average Rsheet:
  - -Pyrex: 320 Ω/sq
  - -Zerodur: 250 Ω/sq

L. Biedermann, T. Beechem, A. Ross, T. Ohta, and S. Howell, *New J. of Physics* 12 (2010) 120516

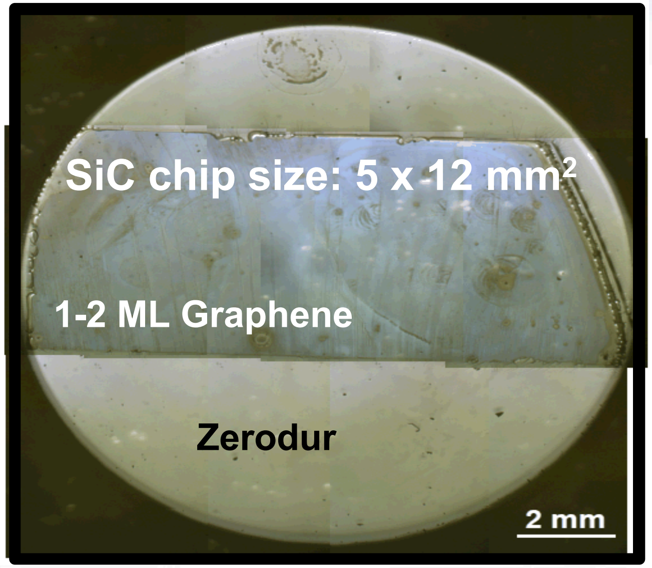
Chip-scale transfer of Si-face graphene to Zerodur

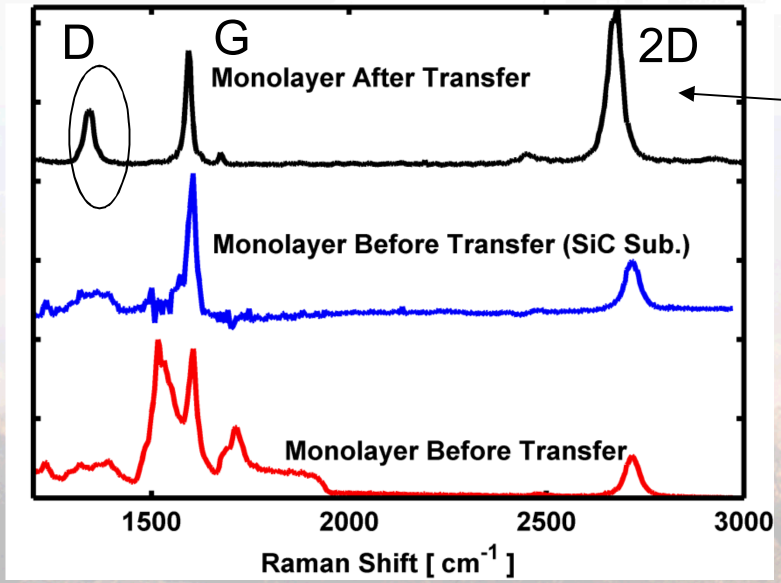


Graphene on SiC, prior to transfer (AFM)



Bright phase = 1 ML
Dark phase = 2 ML





Similar to exfoliated few-layer graphene

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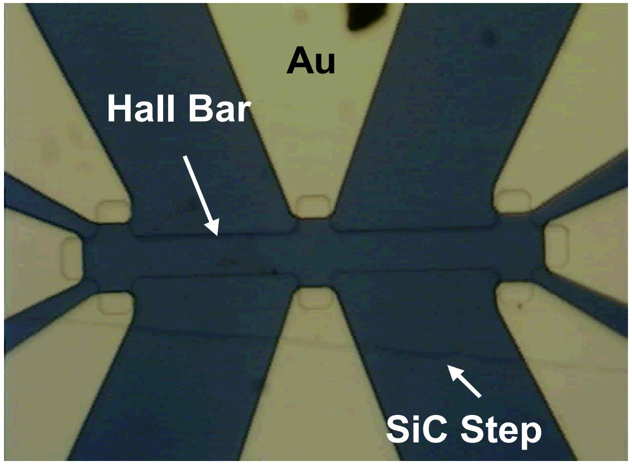
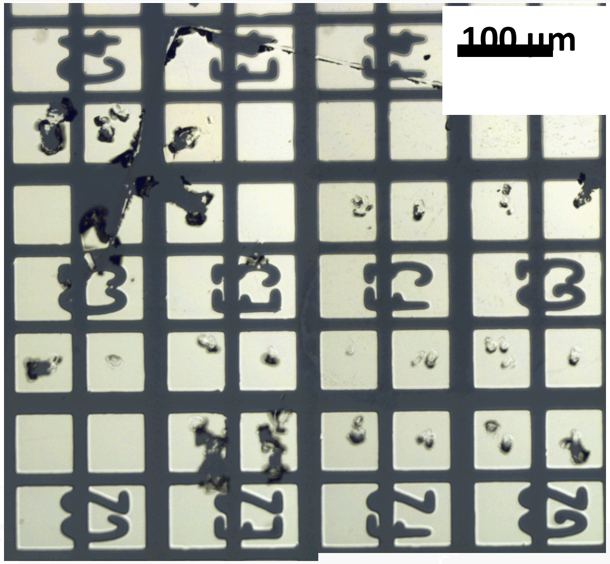
Comparison of transferred Si-face graphene to EG

EG Hall bar on SiC(0001)



EG transferred to Zerodur



- Graphene Rs: ~1630 ± 170 Ω/sq
  - (average from 12 devices)
- EG electron mobility: 14,000 cm^2/Vs
- Electron density: 6×10^{11} cm^{-2}

- Graphene Rs: 1210 ± 260 Ω/sq
- Large area connectivity ~mm^2

Issues:
- Reliability of transfer
- Damage to graphene during transfer and additional processing
- SiC/glass bonding (at some defect locations)

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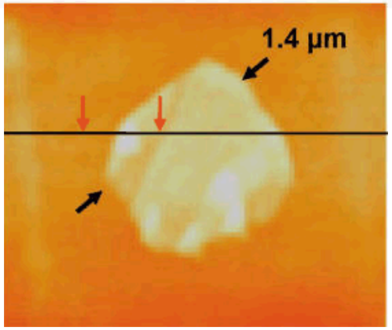
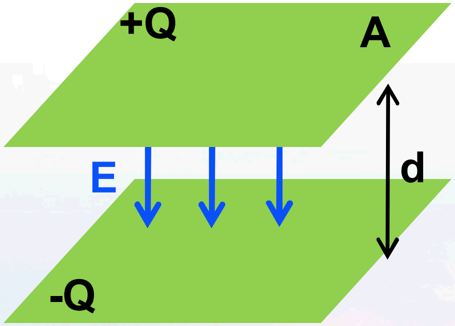
Ongoing work: Electrostatic rather than electrochemical transfer



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).

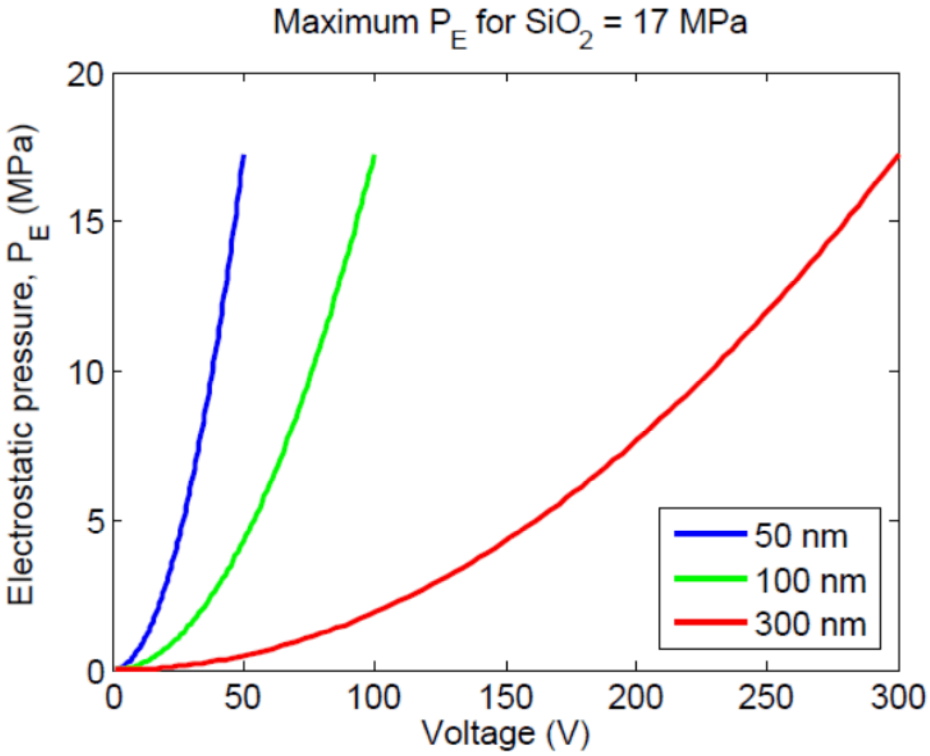
- We are currently developing a method for lower voltage (< 300V) transfer
  - Improving the capacitance of the system for increased electrostatic force



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

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

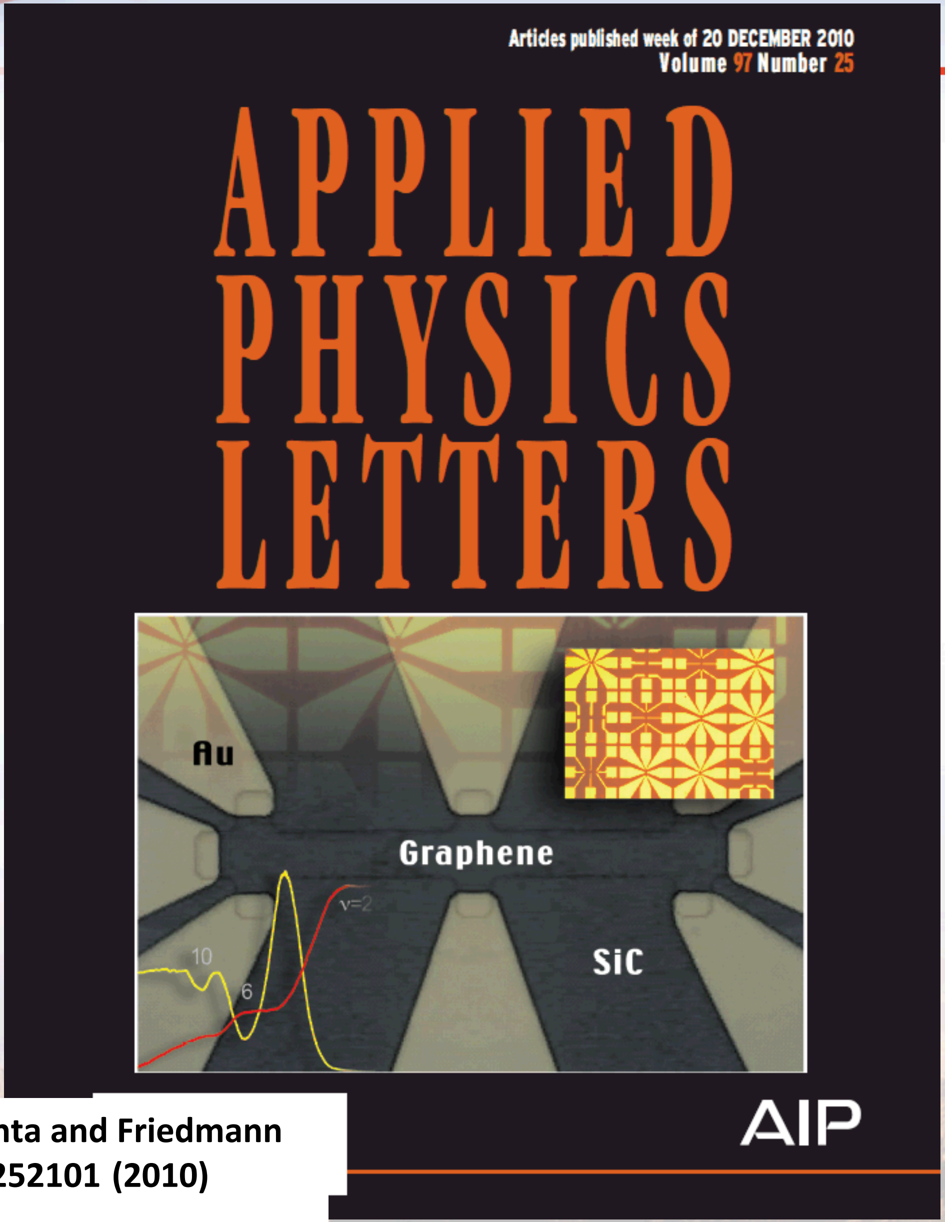
The exfoliation strength of graphite is ~ 0.4 MPa.



Conclusions

- **Demonstrated transfer of C-face EG to Pyrex and Zerodur from SiC**
- **Graphene transferred from C-face was characterized with Raman and micro four probe**
  - C-face graphene can be transferred <u>without inducing defects</u>
  - Average R_s: 320 Ω/Sq (Pyrex) and 250 Ω/Sq (Zerodur)
  - R_s for the transferred graphene was comparable to that for the epitaxial graphene (R_s = 180 ± 70 Ω/sq at ~10 K)
- **Monolayer/Bilayer EG can also be transferred from the Si-face**
  - Increased defects detected by Raman
  - Observed areas of large scale connectivity (~mm^2)
  - average Rs: 1210 ± 260 Ω/Sq
  - Transfer relaxes strain inherent in Si-face EG (data not shown)
- **<u>This technique may be very useful for transferring few layer graphene films prepared on C-Face SiC</u>**

APL Cover

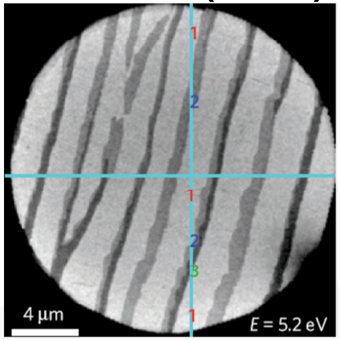


Pan, Howell, Ross, Ohta and Friedmann
Appl. Phys. Lett. 97, 252101 (2010)

Acknowledgements

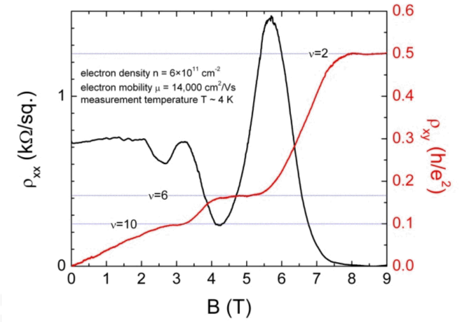
Sandia is in the 3rd year of an internally funded project to develop the scientific basis required for synthesis of high quality graphene

Thermal Decomposition of SiC (Ohta)

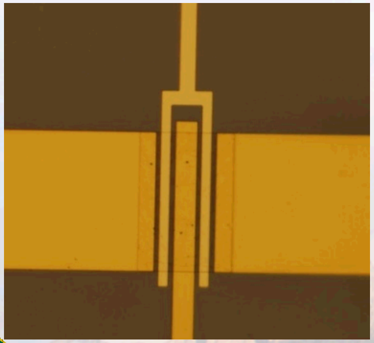


CVD Dep on Metals (McCarty)

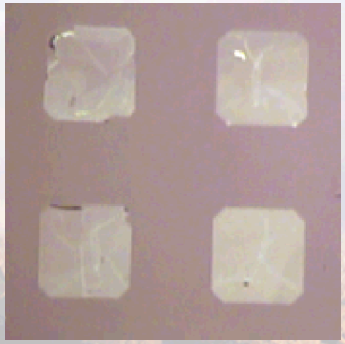
Transport Measurements (Pan, Howell)



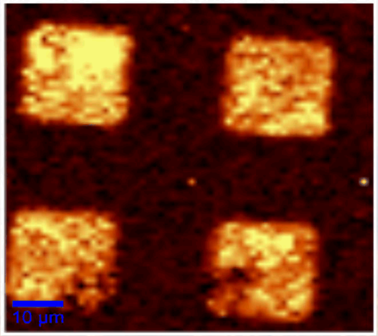
GFET Development (Howell, Ross, Trotter)



Transfer (Biedermann, Howell)



Raman Mapping (Beechem, Ohta)



L. Biedermann, T. Beechem, A. Ross, T. Ohta, and S. Howell, *New J. of Physics* **12** (2010)

Backup Slides

Acknowledgements

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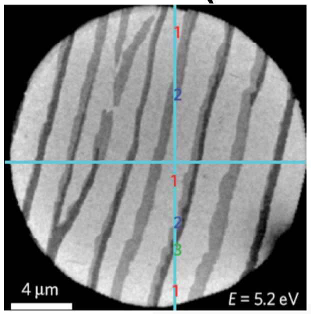
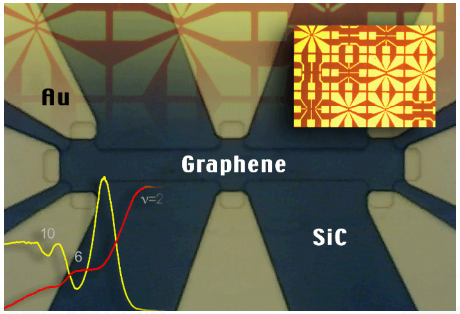
Thermal Decomposition of SiC (Ohta)

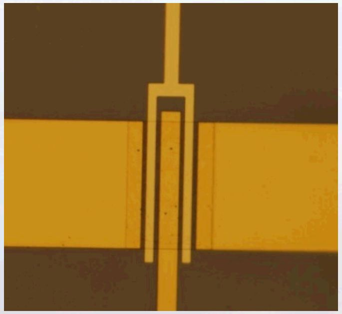


CVD Dep on Metals (McCarty)

Transport Measurements (Pan, Howell, Ross)

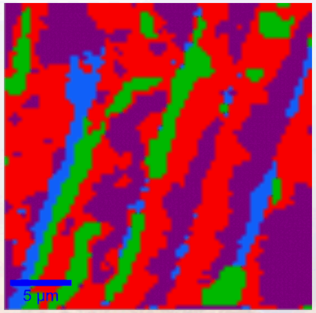


GFET Development (Howell, Ross, Trotter)



Transfer (Biedermann, Howell)

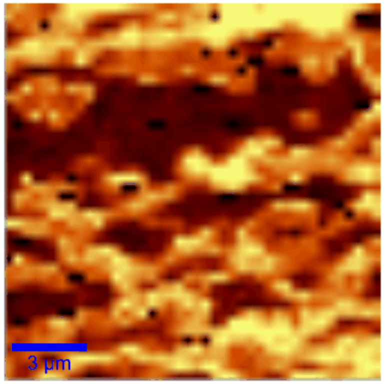
Raman Mapping (Beechem, Ohta)

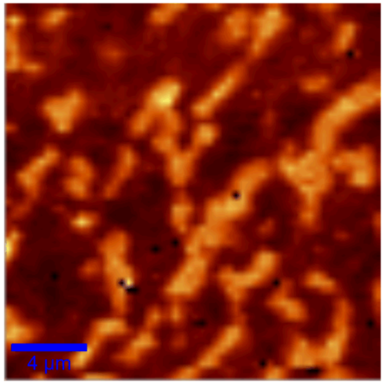


Observation of the integer quantum Hall effect in high quality, uniform wafer-scale epitaxial graphene films
W. Pan, S. Howell, A. Ross, T. Ohta, and T. Friedmann, to be published in *APL,* Dec. 2010 (cover article)

Electrostatic transfer of patterned epitaxial graphene from SiC(000-1) to glass
L. Biedermann, T. Beechem, A. Ross, T. Ohta, and S. Howell, to be published in *New J. of Physics* **12** (2010)

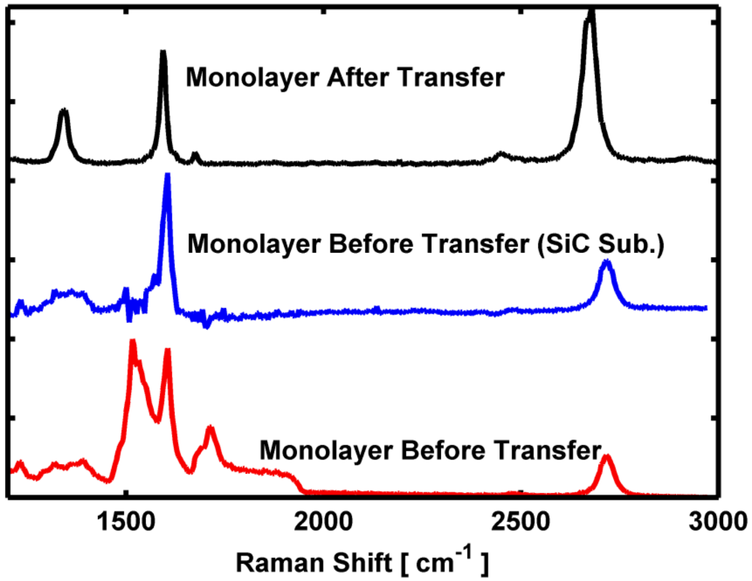
Before (G/SiC) After (G/Zerodur)

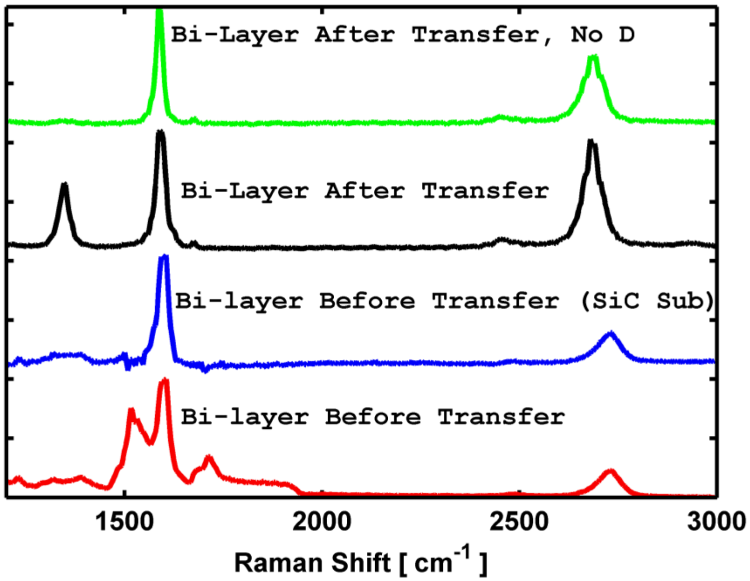




090609 #11 Before After Transfer of Si Face Graphene

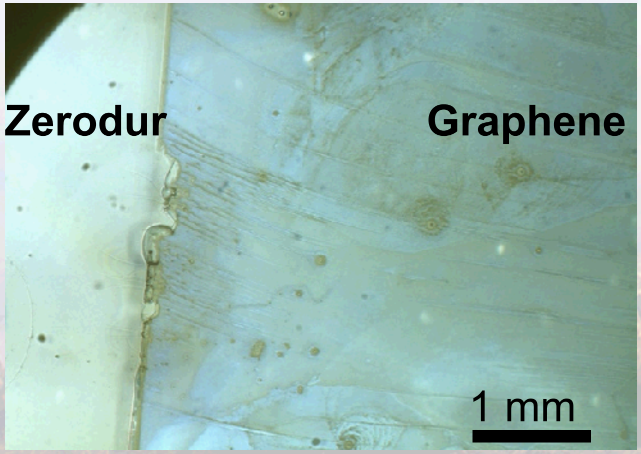
2D-FWHM. Note these are on same scale.



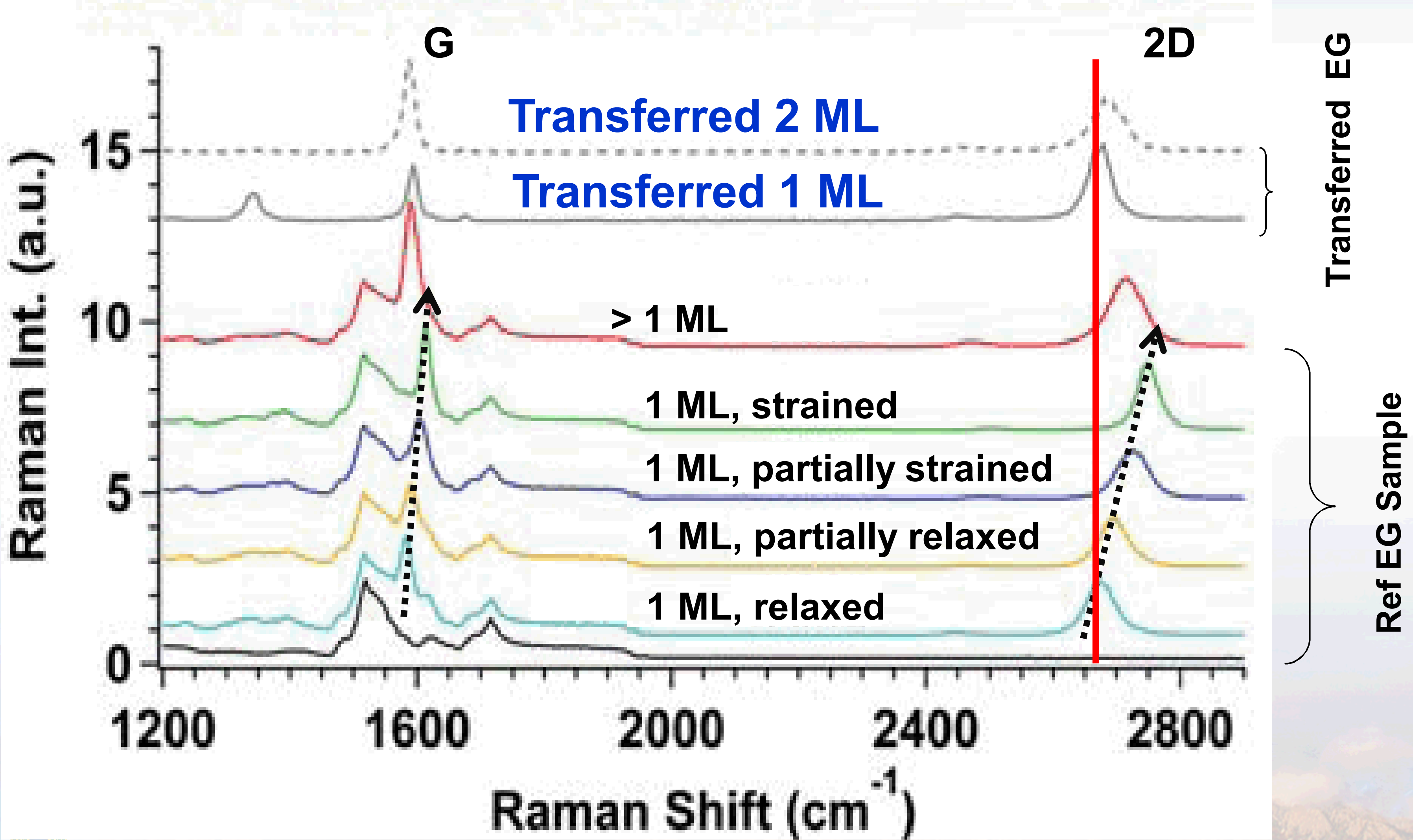


Conclusions

- **Demonstrated transfer of epitaxial graphene to Pyrex and Zerodur from SiC**
- **Graphene transferred from C-face was characterized with Raman and micro four probe**
  - C-face graphene can be transferred without inducing defects
  - The average sheet resistance was 320 Ω/Sq (Pyrex) and 250 Ω/Sq (Zerodur)
  - R_s for the transferred graphene was comparable to that for the epitaxial graphene (R_s = 180 ± 70 Ω/sq at ~10 K)
- **Epitaxial graphene can also be transferred from the Si-face**
  - Transfer relaxes strain inherent in Si-face EG

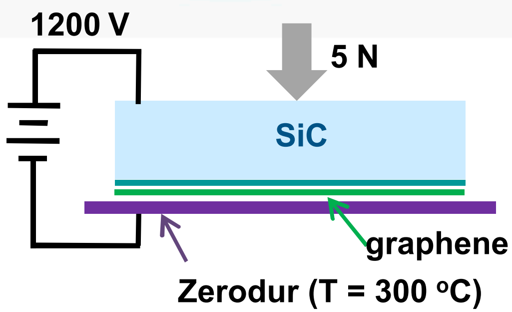


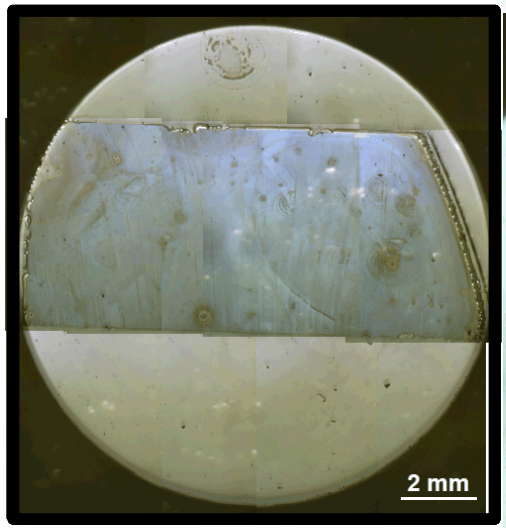
Transfer relaxes strain inherent in epitaxial graphene

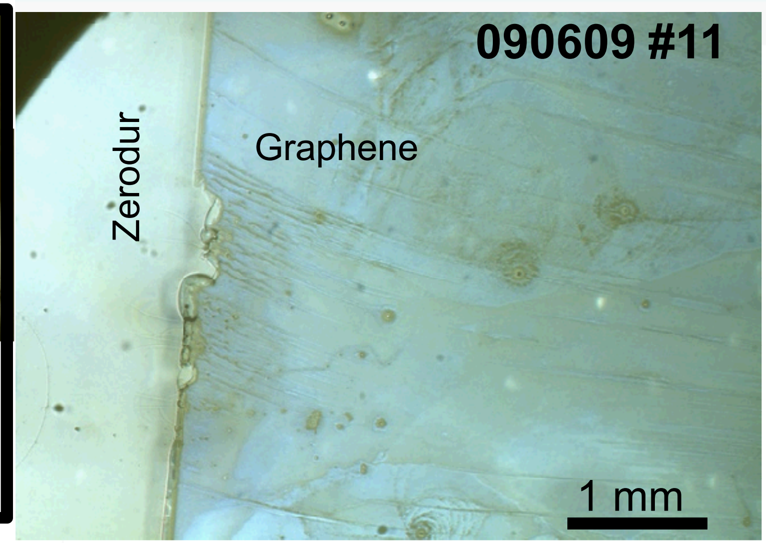


D. Schmidt *et al.*, submitted to *PRB* (2010).

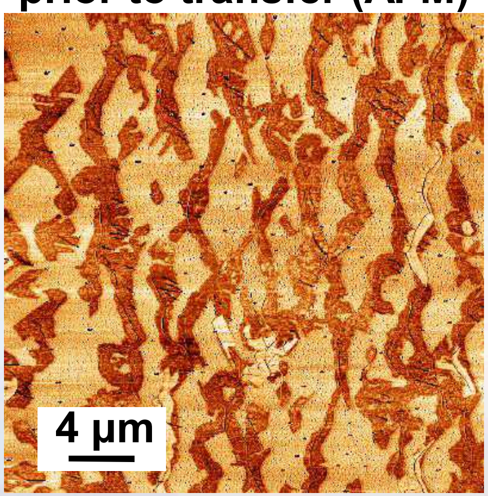
Chip-scale transfer of Si-face graphene to Zerodur







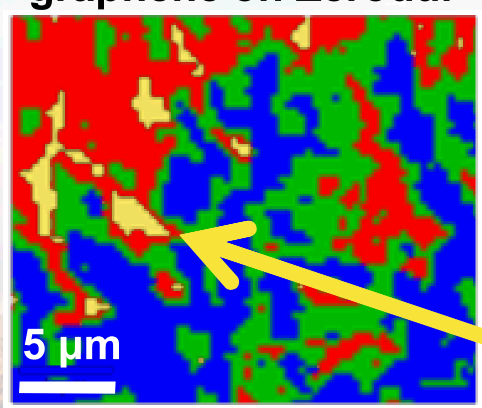
Graphene on SiC, prior to transfer (AFM)

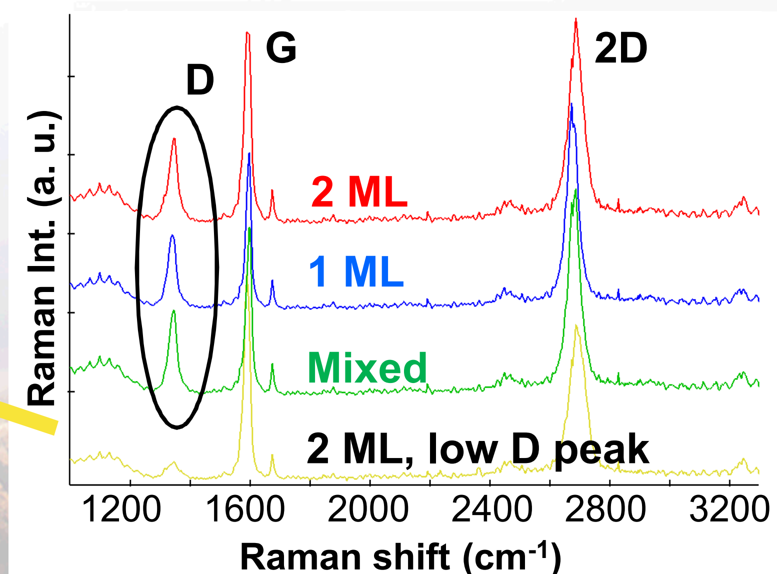


Bright phase = 1 ML

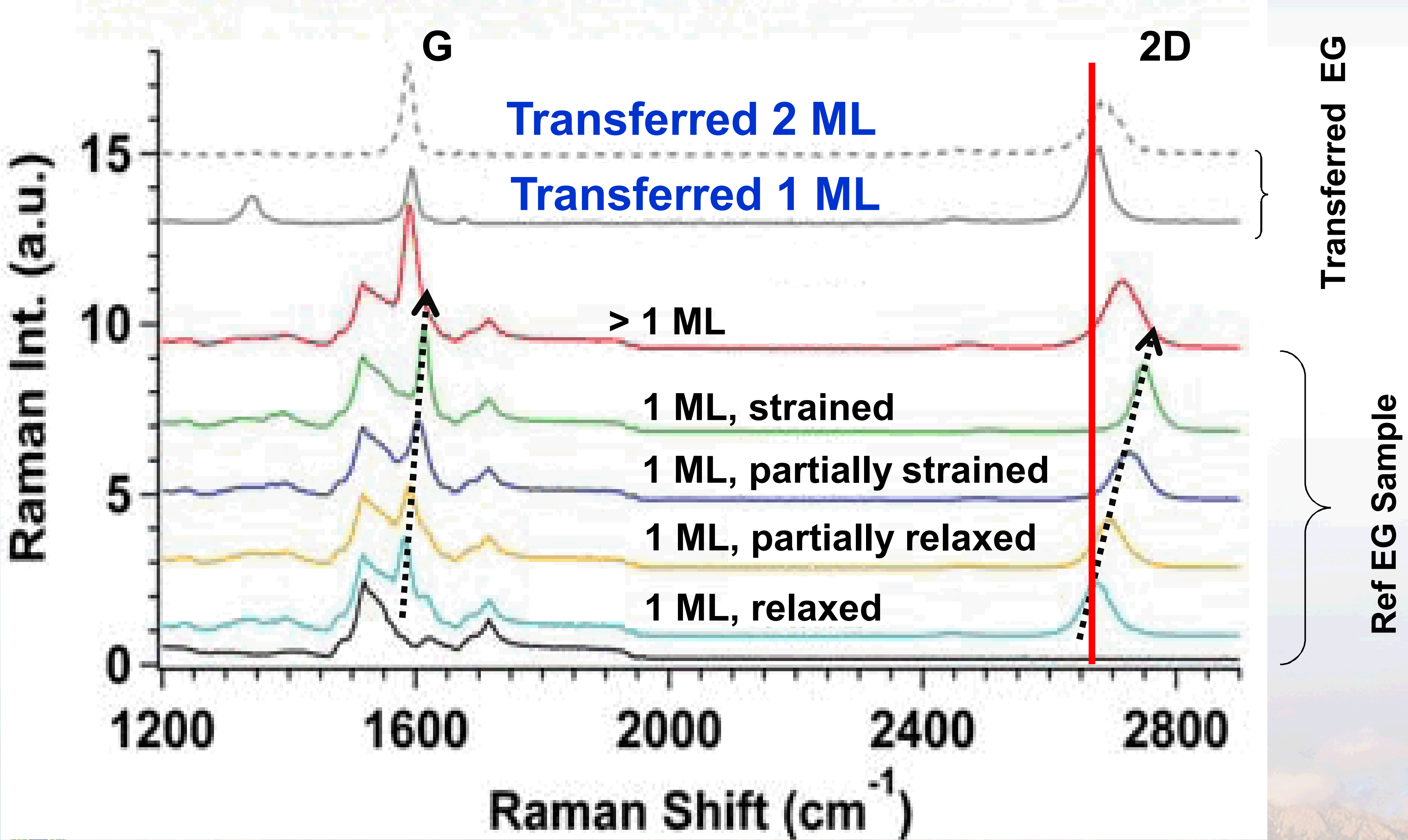
Dark phase = 2 ML

Cluster analysis of graphene on Zerodur



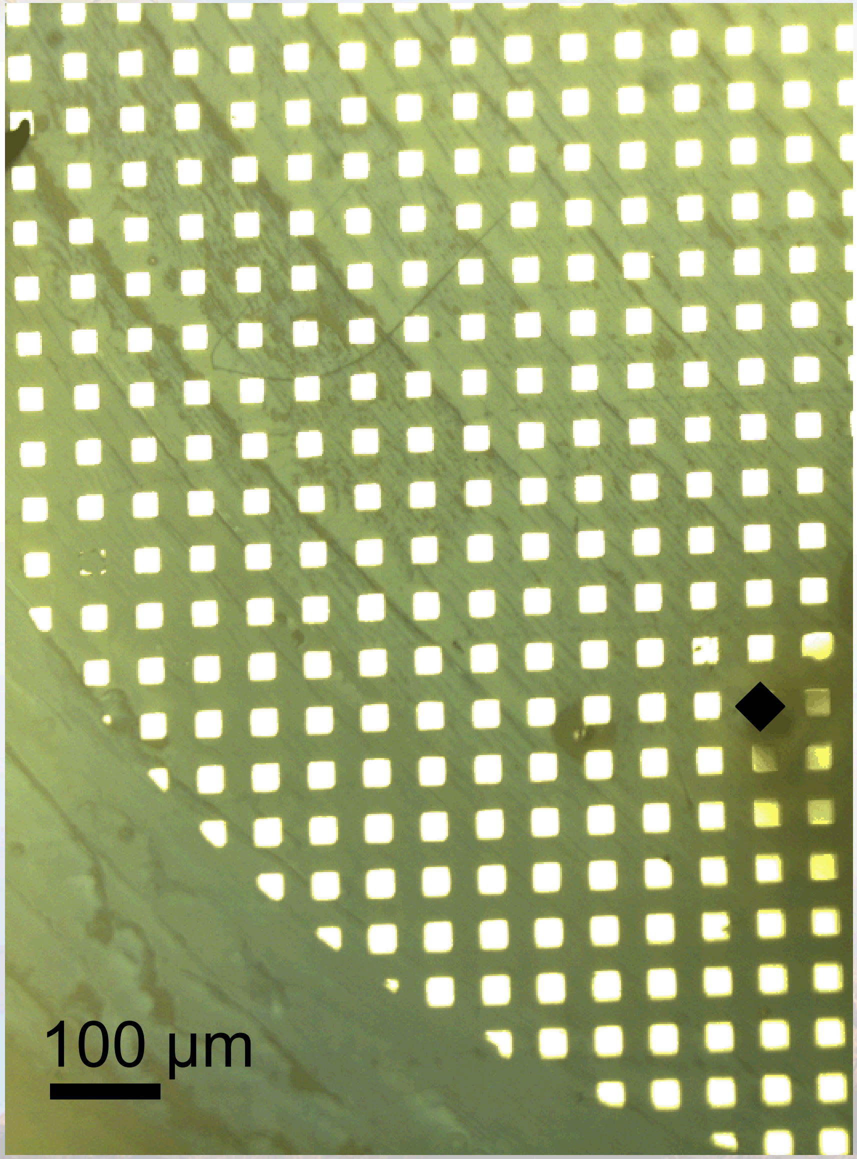


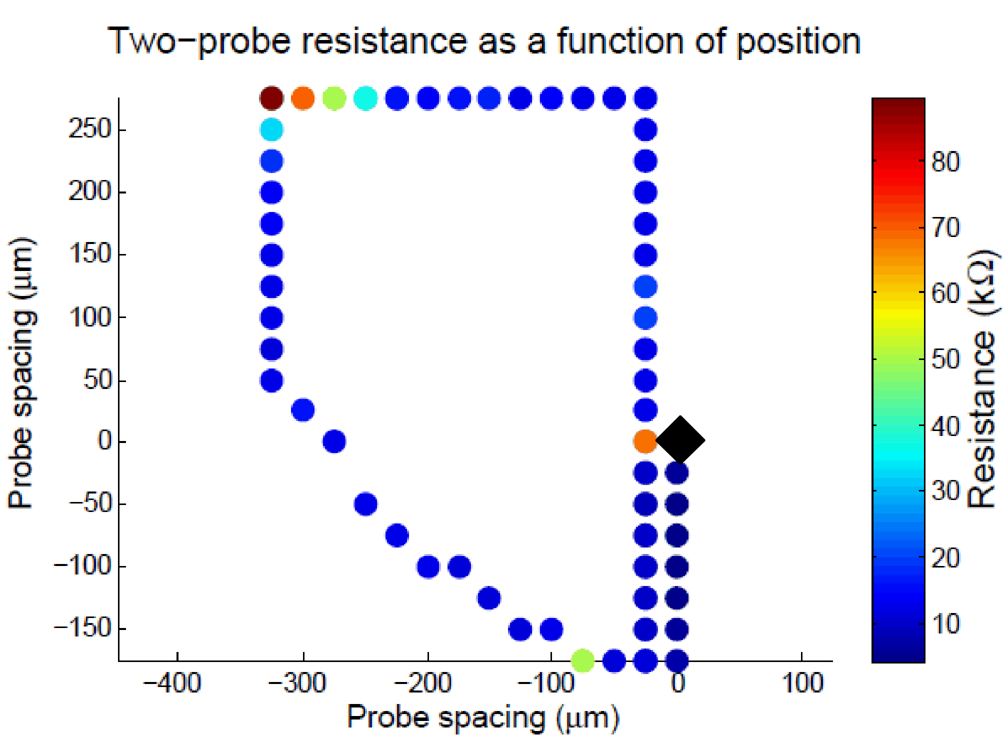
Transfer relaxes strain inherent in epitaxial graphene



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

Transferred Si-face graphene can be conductive over 100s of microns





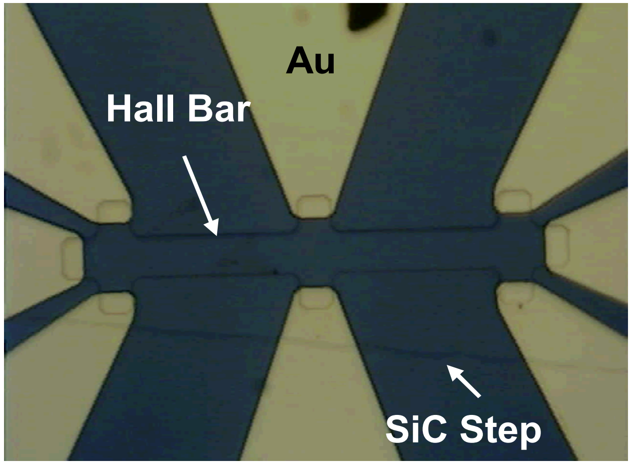
Preliminary four-probe measurements suggest R_s ~ 4 kΩ/sq.

Jim Stanley and Bob Kaplar

Sandia National Laboratories

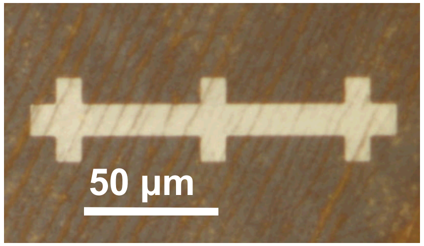
Comparison of transferred Si-face graphene to EG

EG Hall bar on SiC(0001)



- EG electron mobility: 14,000 cm^2/Vs
- Electron density: 6 x 10^{11} cm^{-2}
- Graphene sheet resistance: ~1600 Ω/sq (average from 12 devices)

Hall bars structures, Si-face graphene on Zerodur

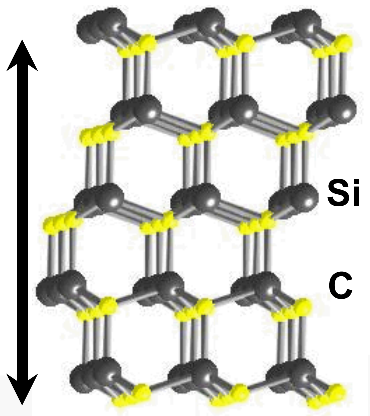


- Poor large scale transfer (most Hall bars are open circuit across their length)
- Measurements across small distances (< 10 µm) do show some electrical activity (~ 5 kΩ -- 5 MΩ)
- The transferred material appears to be isolated ribbons of graphene
  - Breaks could be caused by SiC steps

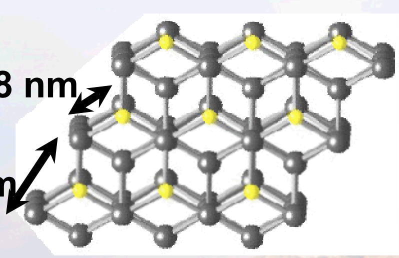
Epitaxial graphene growth on SiC

4H - SiC

Si face (0001)



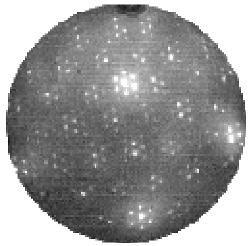
C face (000-1)



1050 °C

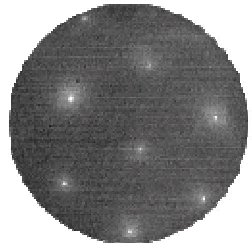
SiC

1150 °C



Buffer layer

1400 °C

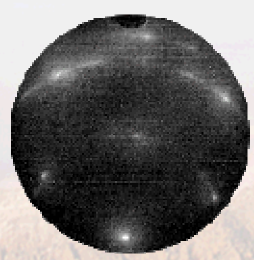


Single-crystal graphite

- 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)

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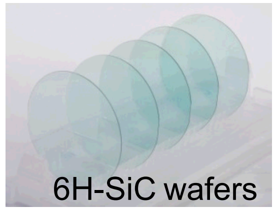
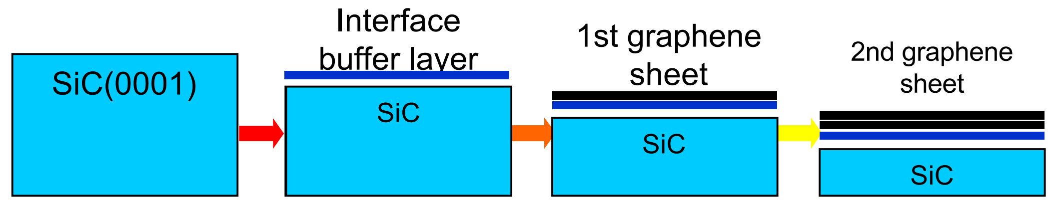
Understanding Graphene Growth on SiC

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





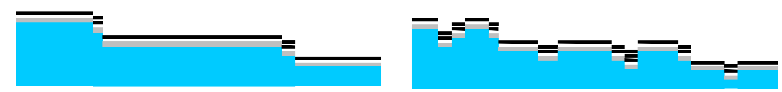
•Argon-assisted graphene formation yields large (~100 μm^2) graphene domains

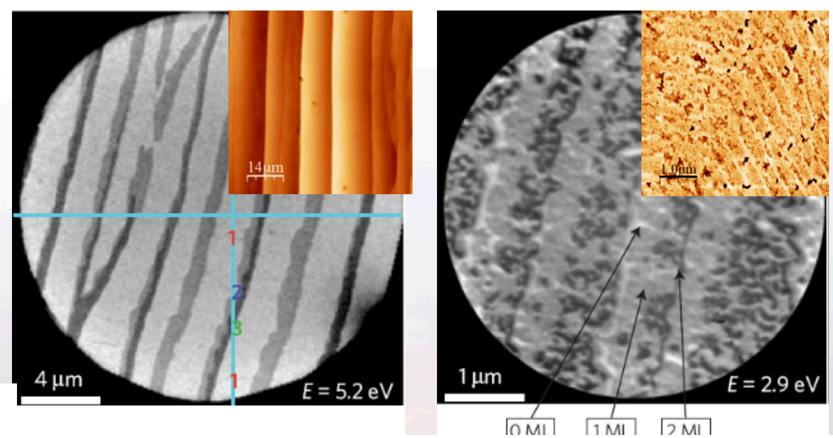
•Samples prepared using Ar atmosphere at atmospheric pressure and high temp

•Using this method we have exquisite control of :

•Domain size

•Percentage coverage of mono/bilayers



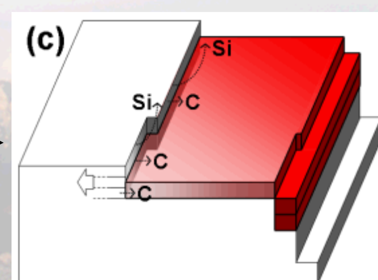


K. V. Emtsev et al., Nature Mater. 8, 203 (2009).
C. Virojanadara et al., Phys. Rev. B 78, 245403 (2008)

•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



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

Ar-mediated synthesis yields high quality epitaxial graphene

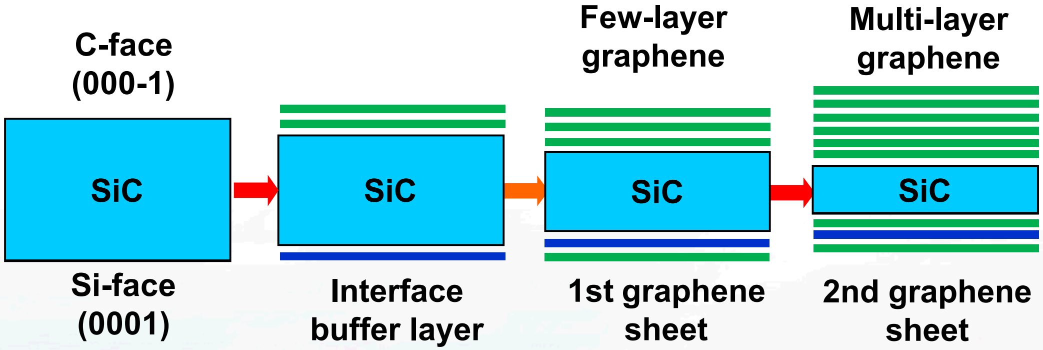
K. V. Emtsev *et al., Nature Mater*. **8**, 203 (2009)..
C. Virojanadara *et al., Phys. Rev. B* **78**, 245403 (2008).
T. Ohta *et al., Phys Rev B* **81** 121411(R) (2010).
Samples provided by T. Ohta

SiC graphitization conditions:

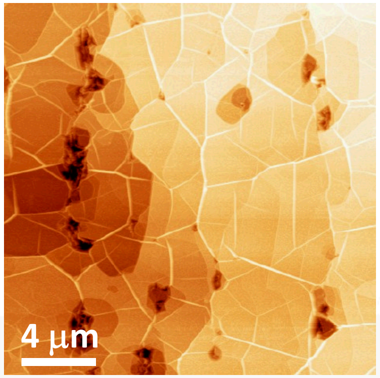
High temperature (>1200 °C)

Argon atmosphere at atmospheric pressure

6H-SiC wafers



C-face (000-1)



Si-face graphene:

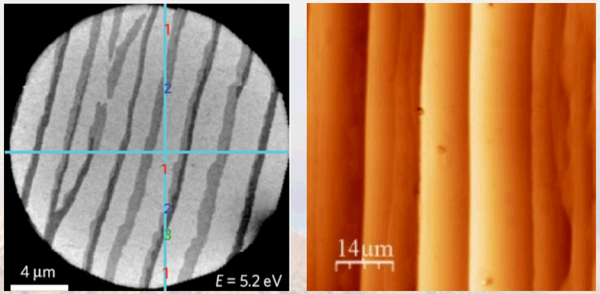
Large (~100 µm^2) graphene domains

μ = 14,000 cm^2/Vs, n = 6 x 10^{11} cm^{-2}

Graphene sheet resistance: ~1600 Ω/sq

IQHE observed on three devices on the same chip

Si-face (0001)



W. Pan, S. Howell, A. Ross, T. Ohta, and T. Friedmann, to be published in *APL*, Dec. 2010

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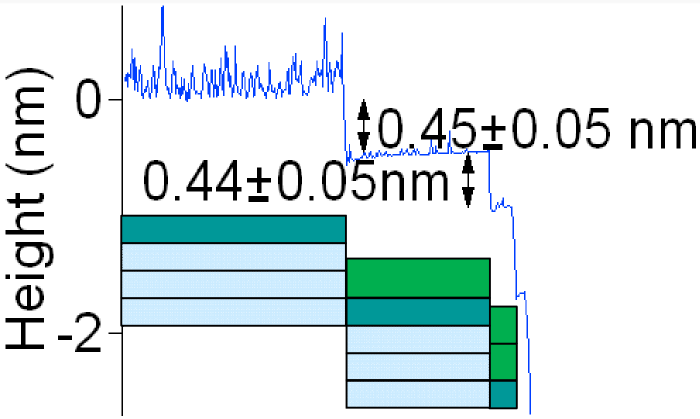
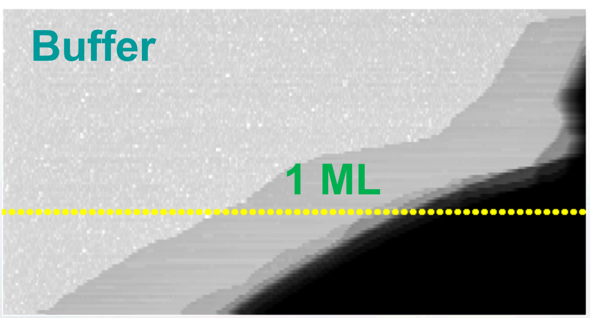
About-face: Si-face epitaxial graphene

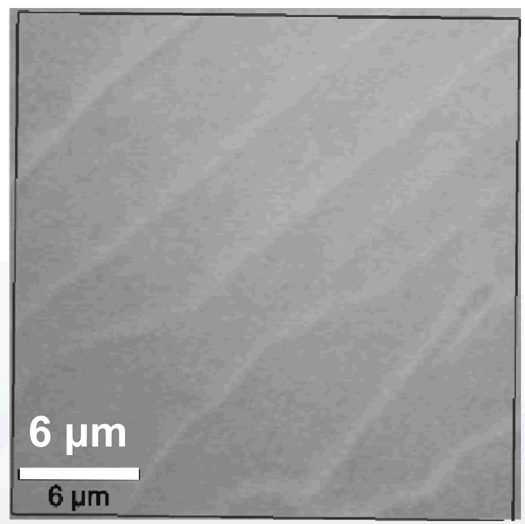




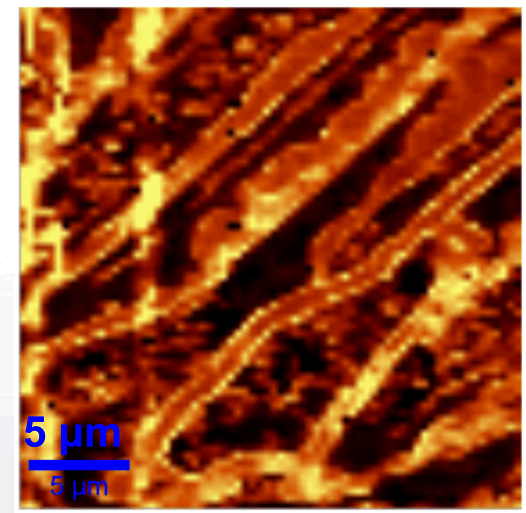
T. Ohta *et al. Phys Rev B* **81,** 121411(R) (2010).

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

Optical



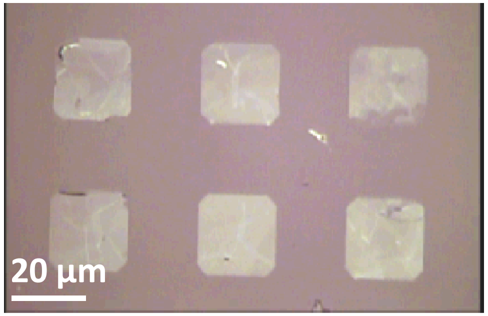
2D FWHM



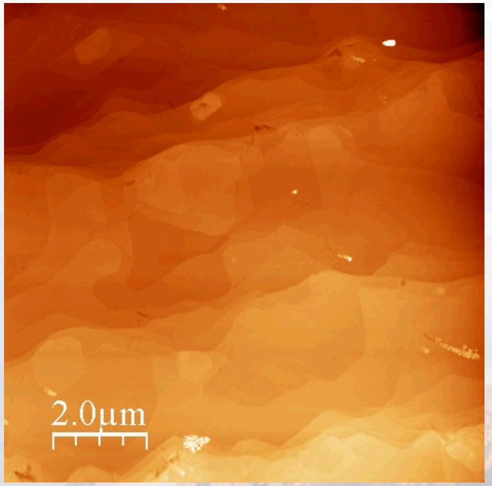
Mobility at 4 K: μ = 14000 cm^2/Vs
Wei Pan, Sandia

How to judge graphene transfer

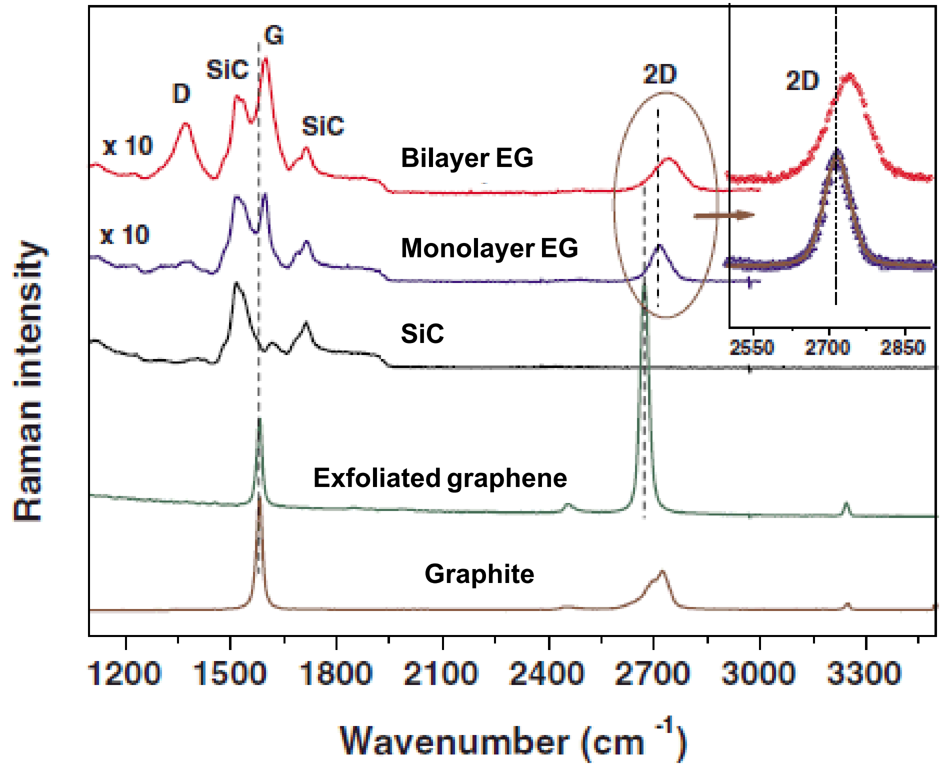
Optical microscopy



Atomic force microscopy



Raman spectroscopy

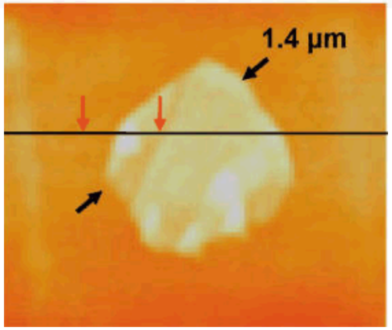


D-band: ~1350 cm^{-1}
G-band: ~1580 cm^{-1}
2D-band: ~2700 cm^{-1}

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

Sandia National Laboratories

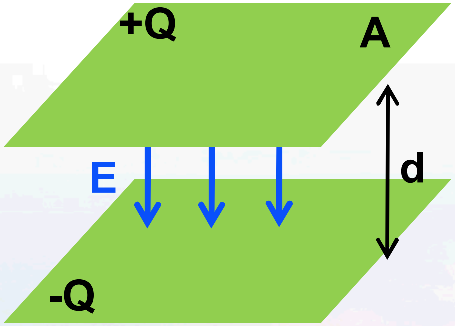
Ongoing work: Electrostatic rather than electrochemical transfer



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

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

- We are currently developing a method for lower voltage (< 30 V) transfer
- We are improving the capacitance of the system for increased electrostatic force



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

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

The exfoliation strength of graphite is ~ 0.4 MPa.