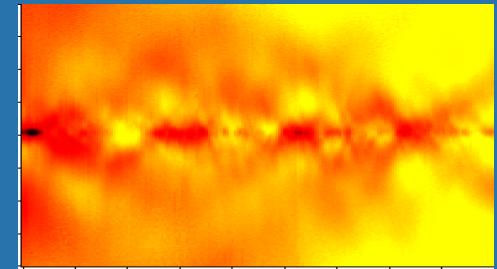
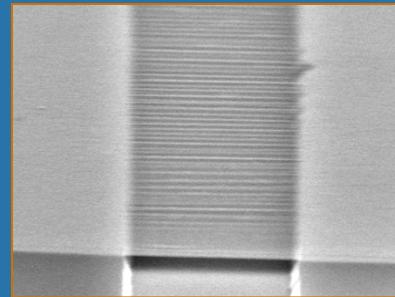
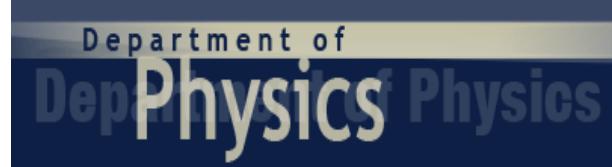
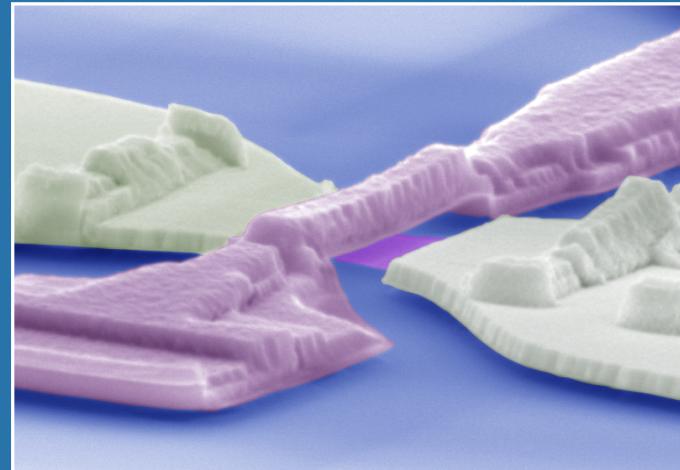


**Chun Ning Lau
(Jeanie)**



**1, 2, 3...
Ripples, Gaps and Transport
in Few-layer Graphene Membranes**



Acknowledgments

Graduate Students



Yongjin Lee



Jhao-wun Huang



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Nathaniel Gilgren



Yanmeng Shi



Petr Stepanov



Son Tran

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Jairo Velasco (Now @ Berkeley)

Hang Zhang (Now @ Caltech)

Lei Jing (Now @ Applied Materials)

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Michael Deo

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Florida Mag Lab

Dmitry Smirnov



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Marc Bockrath



UCR Physics

Chandra Varma



UCR Physics

Vivek Aji



Lancaster

Ed McCann



Tohoku Univ.

Mikito Koshino



UT Austin

Allan MacDonald
Fan Zhang, Jeil Jung



USC

Steve Cronin



Outline

- Suspending Graphene Membranes
- Bilayer Graphene
 - Tunable gap (single particle)
 - Tunable gap (many body interactions)
 - Collective, symmetry-broken states in bilayer graphene
- Trilayer graphene
 - ABA and ABC stacking
 - Tunable interaction-induced gap in ABC-stacked trilayer

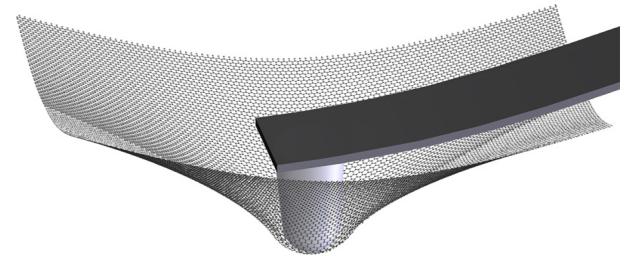
Outline

- Suspending Graphene Membranes
 - free of substrate interference
 - intrinsic optical, mechanical, thermal and electrical properties
- Bilayer Graphene
 - Tunable gap (single particle)
 - Tunable gap (many body interactions)
 - Collective, symmetry-broken states in bilayer graphene
- Trilayer graphene
 - ABA and ABC stacking
 - Tunable interaction-induced gap in ABC-stacked trilayer

Strange Graphene Mechanics

Graphene is very strong and tough

Breaking strength is ~ 200 times of steel



Source: Columbia Univ.

Graphene is very elastic

can be stretched by 25% and return to its original shape
(most materials can only be stretched by one-tenth of 1 percent)

Graphene is very soft

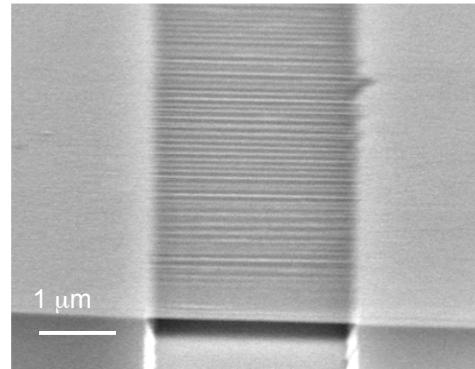
Single atomic layer, bends or
buckles very easily

Mechanical Properties: Periodic Ripple Formation

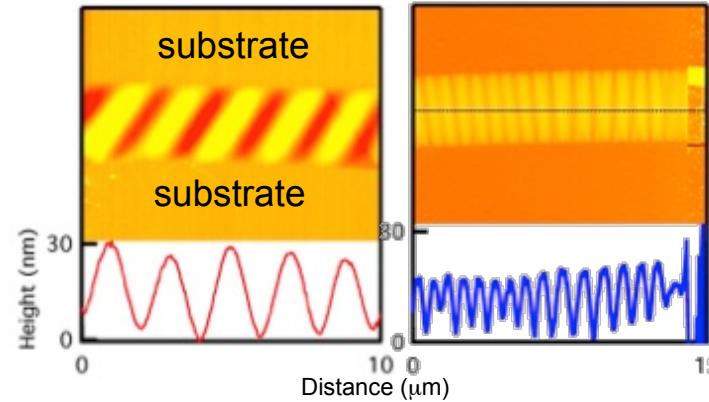
Directly exfoliate graphene sheets across pre-defined trenches

- Many graphene sheets are not flat, but spontaneously form ripples
- Almost perfectly sinusoidal profile
 - ◆ thickness: 0.3 nm (single layer) -- 16 nm
 - ◆ amplitude: 0.7 to 30 nm
 - ◆ wavelength: 370 nm – 5 μm

SEM



AFM



Graphene as an Elastic Membrane

VOLUME 90, NUMBER 7

PHYSICAL REVIEW LETTERS

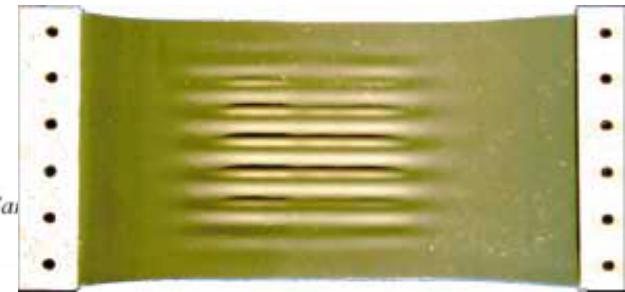
week ending
21 FEBRUARY 2003

Geometry and Physics of Wrinkling

E. Cerdá^{1,2} and L. Mahadevan^{1,*}

¹*Department of Applied Mathematics and Theoretical Physics, University of Cambridge,
Silver Street, Cambridge CB3 9EW, United Kingdom*

²*Departamento de Física, Universidad de Santiago de Chile, Avenida Ecuador 3493, Casilla 307, Correo 2, Sa-*
(Received 25 June 2002; published 19 February 2003)



- competition between bending and stretching
- ripples induced by longitudinal strains or shears

$$\frac{A\lambda}{L} = \sqrt{\frac{8\nu}{3(1-\nu^2)}}t$$

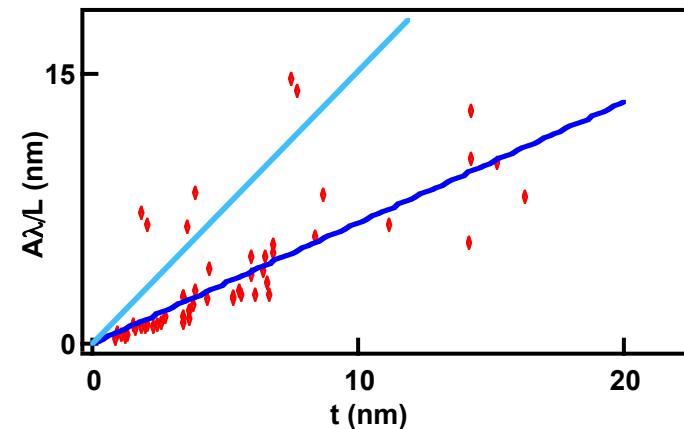
in-plane shear

$$\frac{A\lambda}{L} = \sqrt{\frac{8}{3(1+\nu)}}t$$

in-plane strain

A=amplitude, L=length, λ =wavelength,
 t =thickness, ν =Poisson ratio~0.165

Data from 51 samples



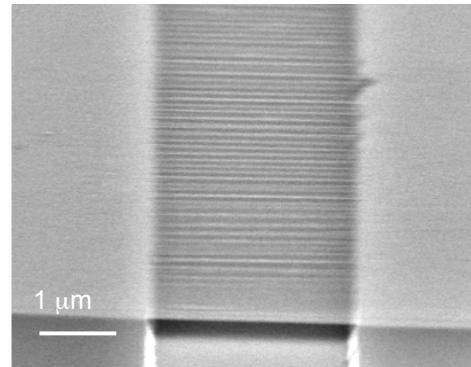
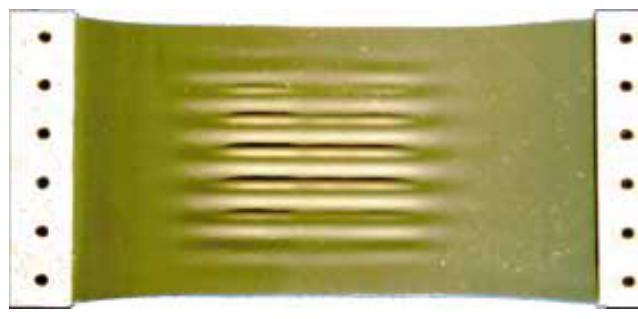
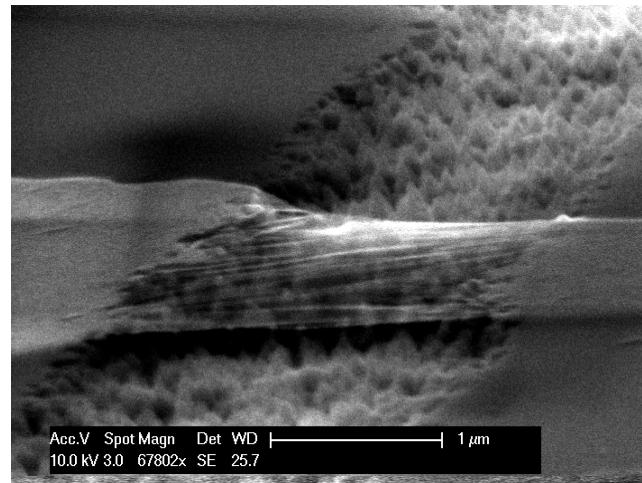
W. Bao, F. Miao, Z. Chen, H. Zhang, W. Jang, C. Dames, and C. N. Lau, Nature Nanotechnology, 4, 562 (2009).

Graphene as the World's thinnest Saran Wrap

macroscopic

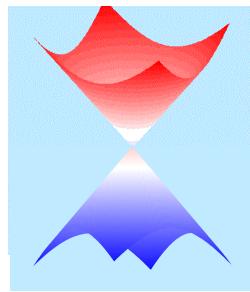
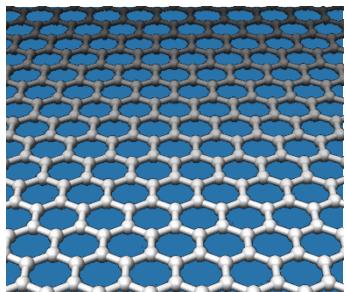


mesoscopic



Graphene's Double Identity

Extraordinary Conductor

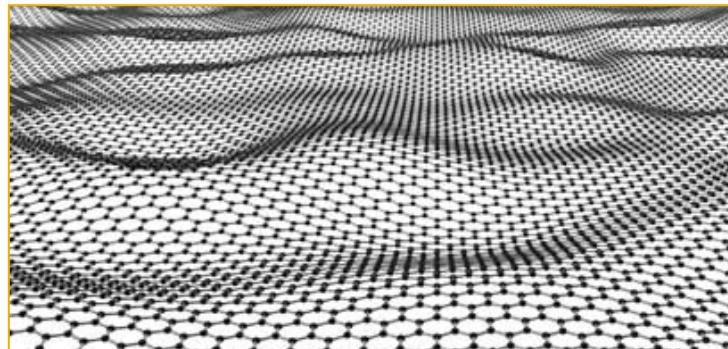


New model system for condensed matter research and electronic materials

Linear dispersion, tunable carrier, surface 2DEG, high thermal and electrical conductivity

....

2D Elastic Membrane



Thinnest isolated membrane with exceptional mechanical properties

Castro Neto, Guinea,
Katsnelson, Brey, Louie, etc

Exploit Electrical Properties of Rippled Graphene?
superlattices, strain-based engineering...

Engineering Based on Strain and Ripples

All-graphene integrated circuits via strain engineering

Vitor M. Pereira, Antonio H. Castro Neto

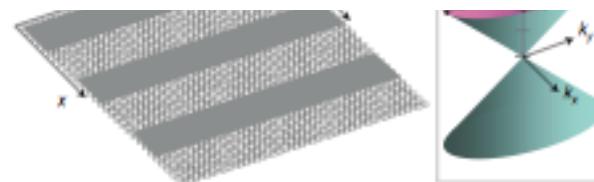
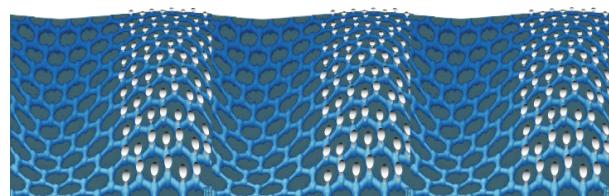
(Submitted on 27 Oct 2008 (v1), last revised 19 Feb 2009 (this version, v3))

We propose a route to all-graphene integrated electronic devices by exploring the influence of strain on the electronic structure of graphene. We show that strain can be easily tailored to generate electron beam collimation, 1D channels, surface states and confinement, the basic elements for all-graphene electronics. In addition this proposal has the advantage that patterning can be made on substrates rather than on the graphene sheet, thereby protecting the integrity of the latter.

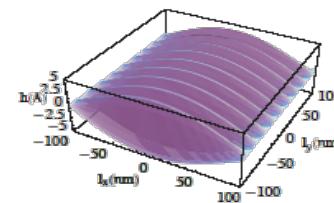
NOVIKOV & LEVKOV, TRL (2008)

Brey & Fertig, arxiv (2009).

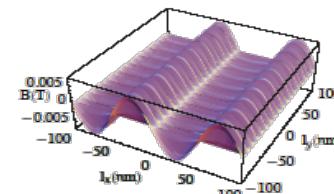
- Modified band structure
- Anisotropic transport
- Supercollimation
- Inducing effective magnetic field
- Selective sp^2 - sp^3 modification



Park, Yang, Son, Cohen, Louie, *Nature Physics* (2008)



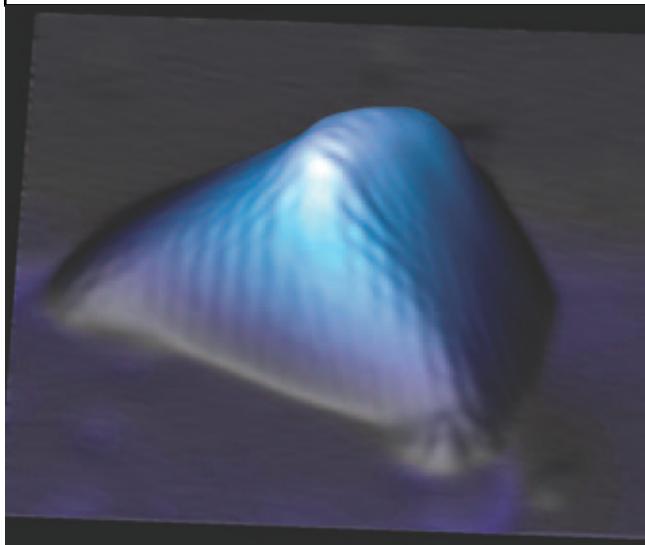
Paco Guinea,
Katsnelson and co.



Engineering Based on Strain and Ripples

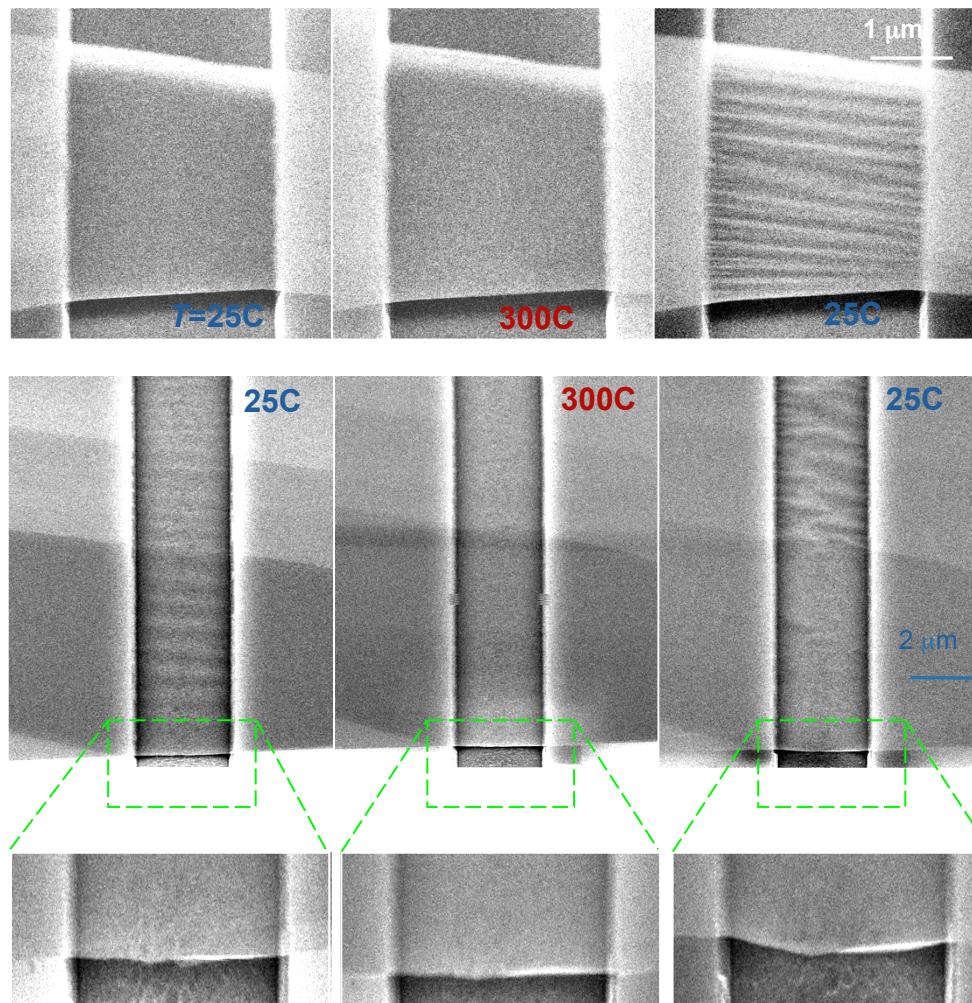
Strain-Induced Pseudo-Magnetic Fields Greater Than 300 Tesla in Graphene Nanobubbles

N. Levy,^{1,2*}† S. A. Burke,^{1*}‡ K. L. Meaker,¹ M. Panlasigui,¹ A. Zettl,^{1,2} F. Guinea,³
A. H. Castro Neto,⁴ M. F. Crommie^{1,2§}



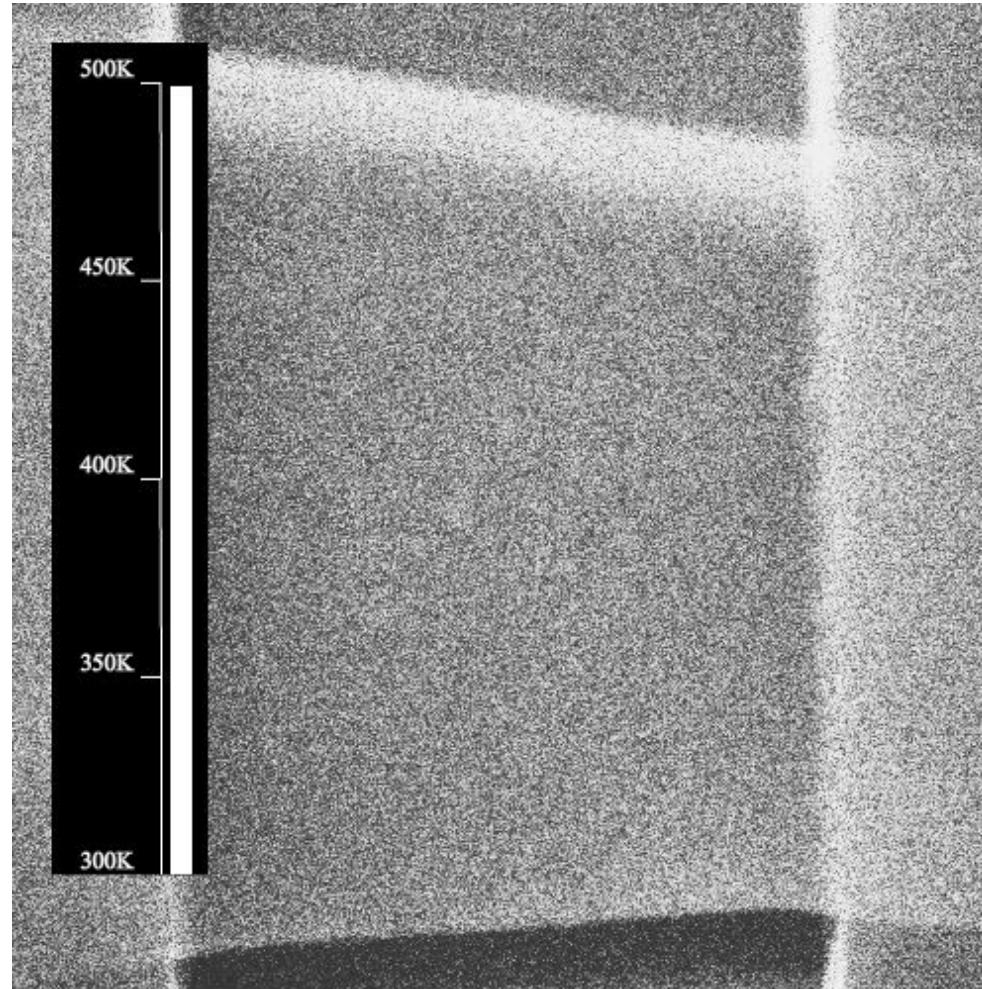
Highest steady magnetic field on earth:
 $= 45\text{T}$
in National High Magnetic Field Lab,
Tallahassee, FL.

In Situ SEM imaging of annealing effect

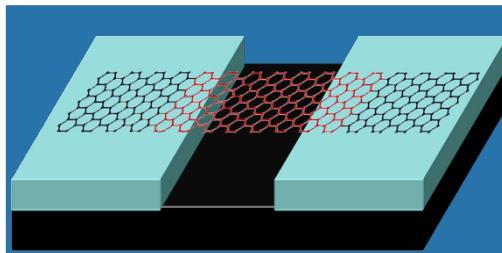


- Ripples have larger wavelengths and amplitudes
- Membranes buckle upward or towards the bottom of the trench

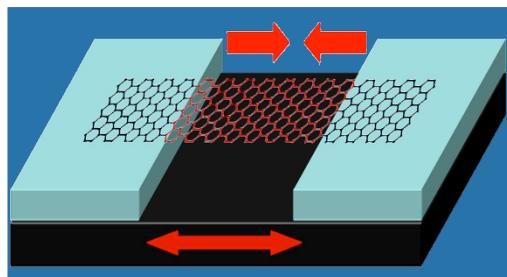
Movie of ripple formation



Mechanism of ripple formation

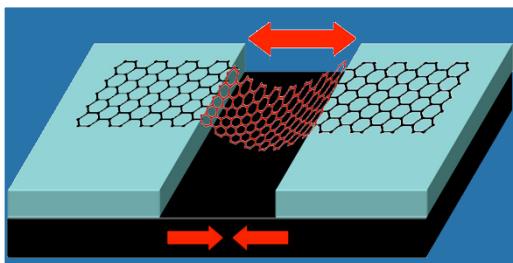


Graphene has a *negative* thermal expansion coefficient



Heating

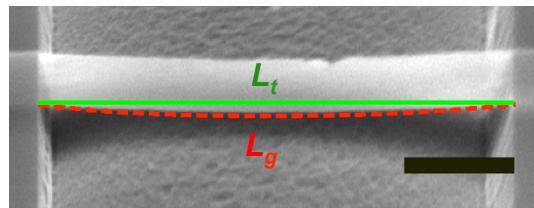
graphene contracts, substrate expands
→ erasing pre-existing ripples



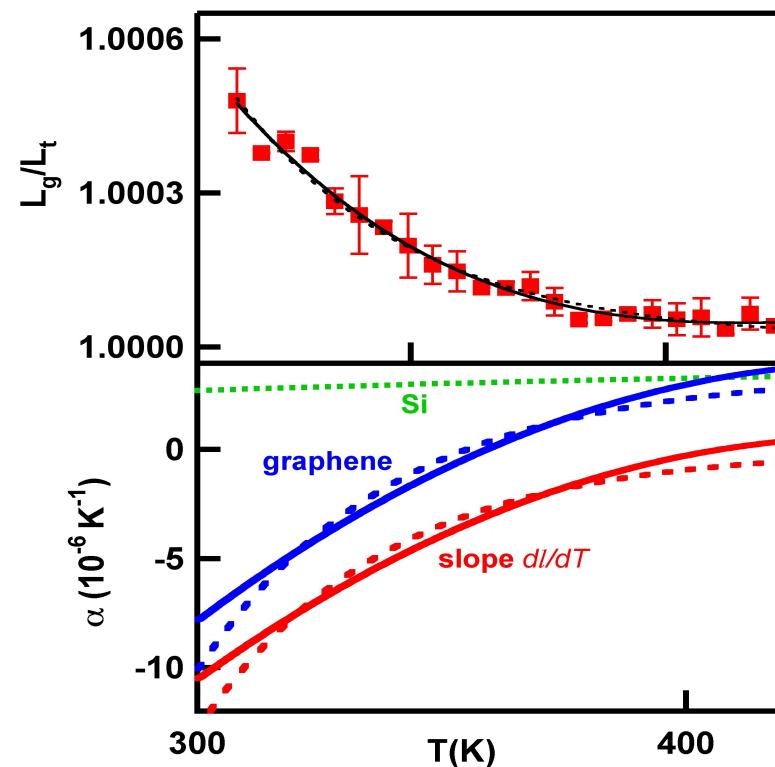
Cooling

- graphene expands, substrate contracts
- bending is easier than sliding
- edges remain pinned by the trench edges
- ripples (transverse)
slacks (longitudinal)

Measurement of Thermal Expansion Coefficient



- Single layer graphene heated to 500 K and cools down slowly
- Compute $I(T)=L_g(T)/L_t(T)$ at different temperatures
- Slope $b = \frac{dl}{dT} \approx \alpha - \alpha_{Si}$
- Important for graphene synthesis and thermal management of graphene electronics

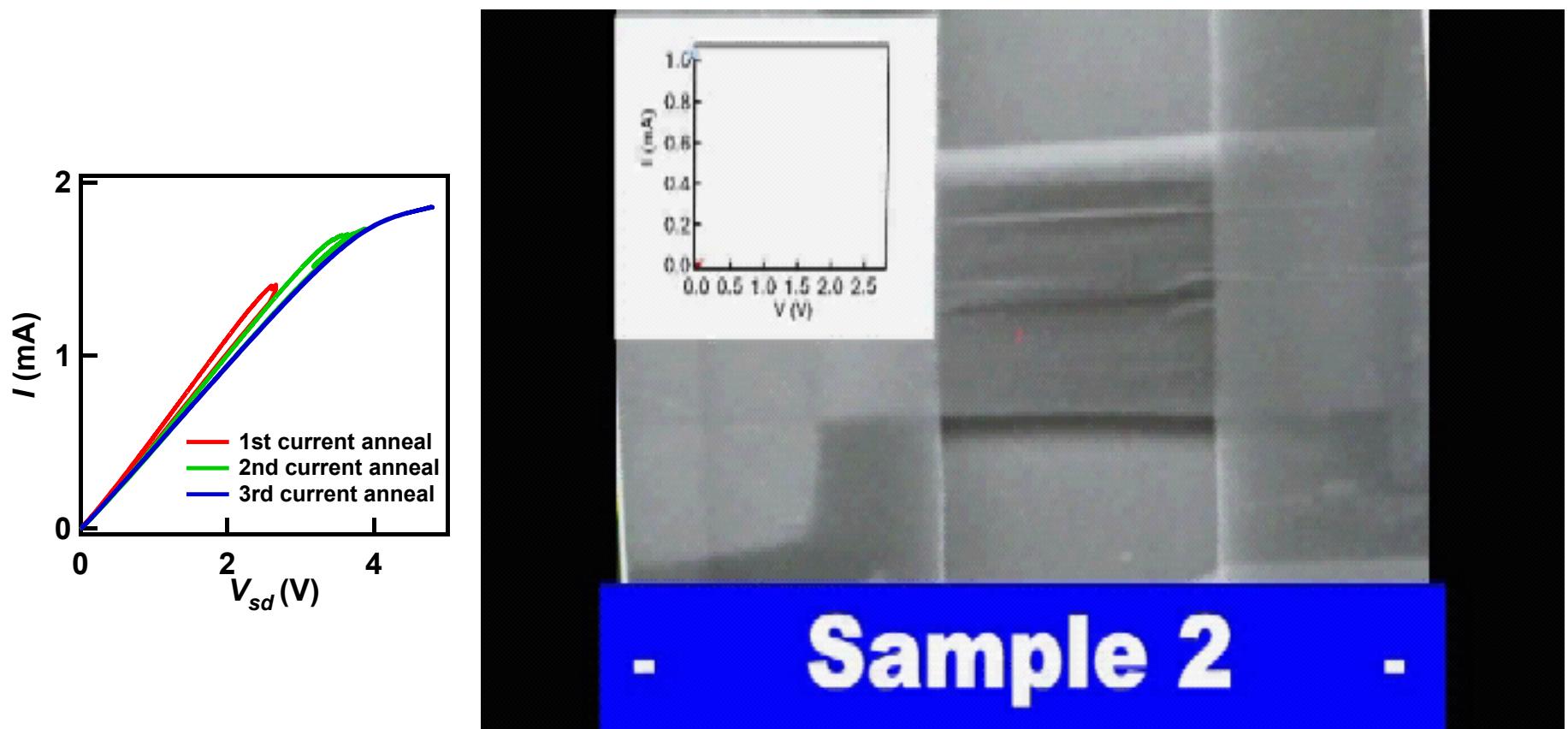


W. Bao, F. Miao, Z. Chen, H. Zhang, W. Jang, C. Dames, and C. N. Lau, Nature Nanotechnology, 4, 562 (2009).

Current Annealing

- Current annealing performed at 4K
- Optimal when current saturates, $\sim 0.2\text{mA}/\text{layer}/\mu\text{m}$

Moser et al, APL.



H. Zhang, W. Bao, Z. Zhao, J.-W. Huang, B. Standley, G. Liu, F. Wang, P. Kratz, L. Jing, M. Bockrath, C. N. Lau,
Nano Lett., 12, 1772 (2012)

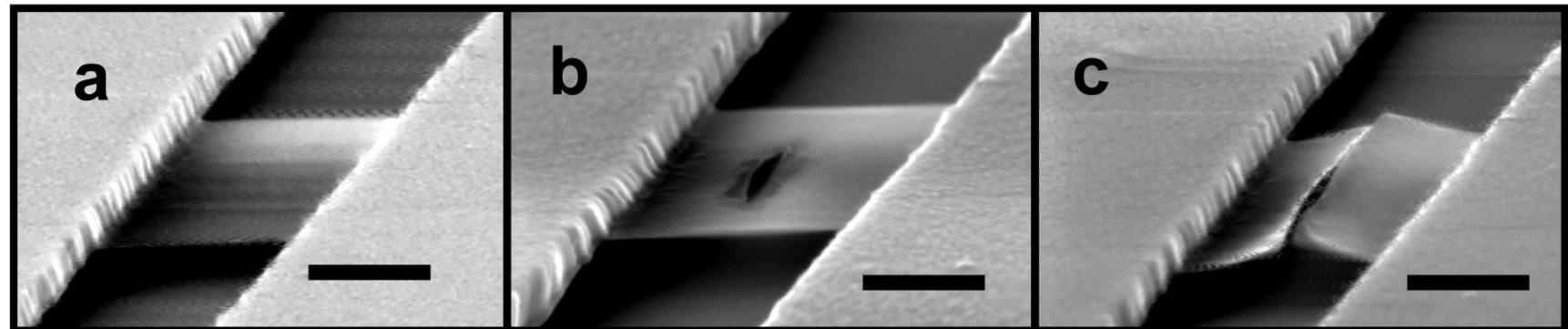
In situ Imaging of Annealing and Electromigration

Further annealing

→ Ripple starts to form due to negative thermal expansion of graphene

→ abrupt changes in I-V curves

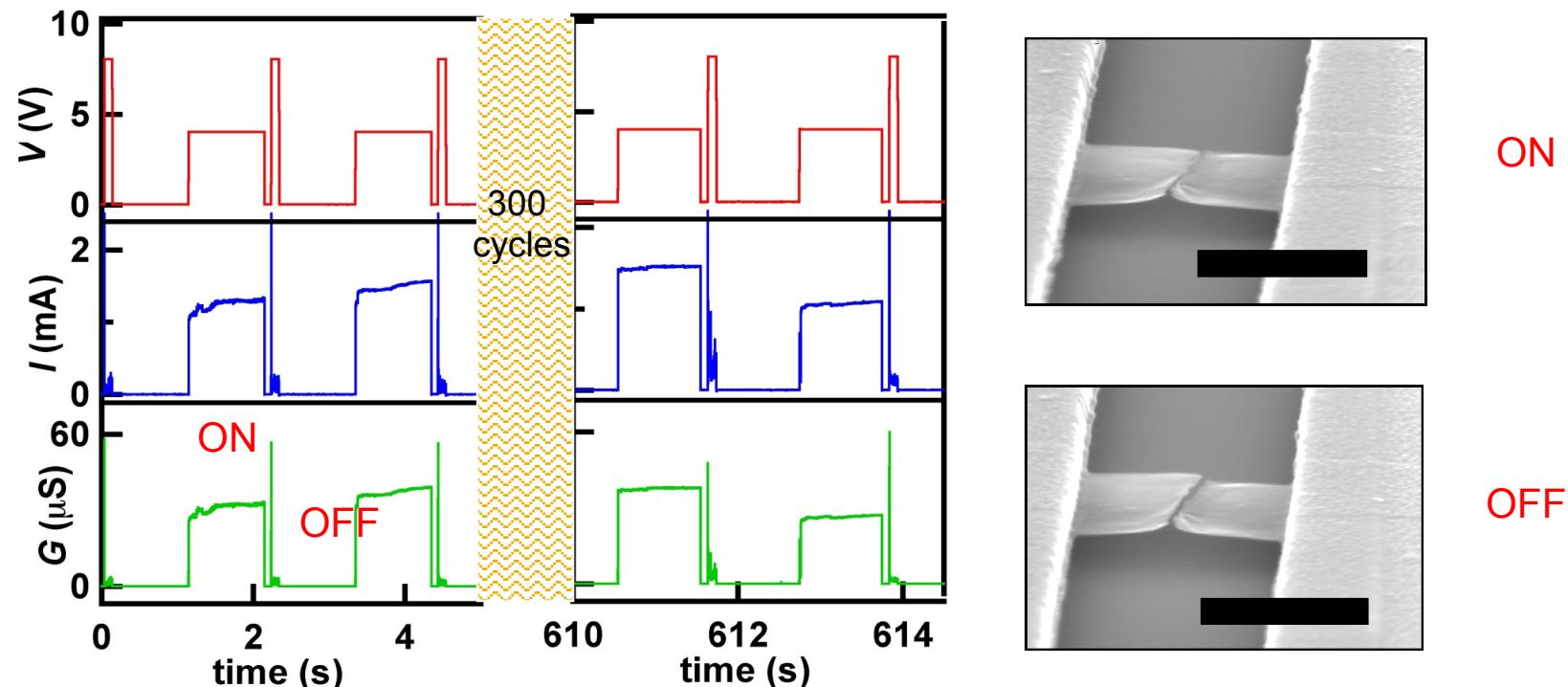
→ Electromigration occurs



H. Zhang, W. Bao, Z. Zhao, J.-W. Huang, B. Standley, G. Liu, F. Wang, P. Kratz, L. Jing, M. Bockrath, C. N. Lau,
Nano Lett., 12, 1772 (2012)

Atomic Switches in Suspended Graphene

- After electromigration → atomic switches
- Reversible, cyclable switching
- Proposed mechanism: formation and dissolution of carbon chains

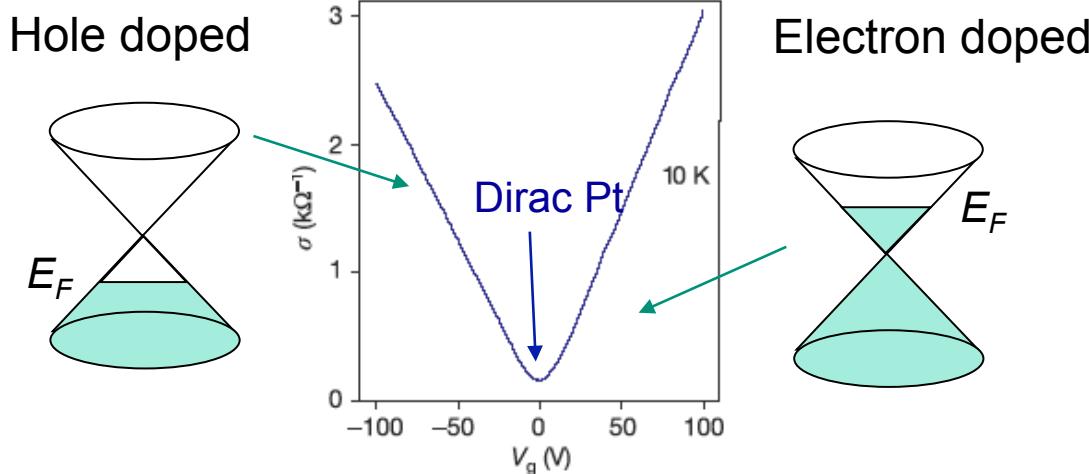


H. Zhang, W. Bao, Z. Zhao, J.-W. Huang, B. Standley, G. Liu, F. Wang, P. Kratz, L. Jing, M. Bockrath, C. N. Lau,
Nano Lett., 12, 1772 (2012)

Outline

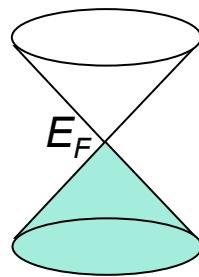
- Suspending Graphene Membranes
- Band gap engineering in Bilayer Graphene
 - Tunable gap (single particle)
 - Tunable gap (many body interactions)
 - Collective, symmetry-broken states in bilayer graphene
- Trilayer graphene
 - ABA and ABC stacking
 - Tunable interaction-induced gap in ABC-stacked trilayer

Graphene Bipolar FET



Novoselov et al, *Science*, 2004.

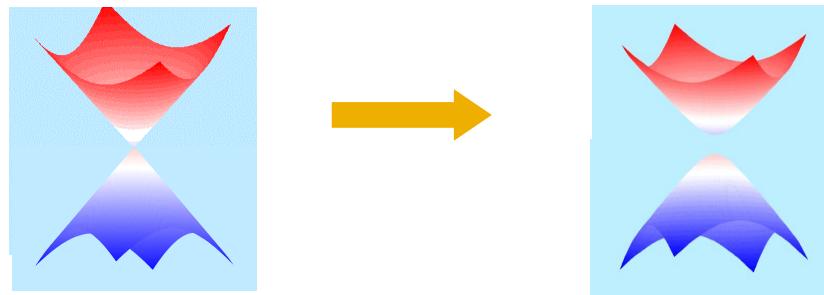
Miao et al, *Science*, 2007.



- Conductivity increases (more or less) linearly with charge density n $\sigma \propto V_g \propto n$
- σ remains finite at Dirac point $\rightarrow \sigma_{\min}$
(Novoselov et al, *Nature*, 2005, Zhang et al, *Nature* 2005, Lau group, *Science* 2007; Kim group, Fuhrer group, Das Sarma group, Ando, Castro Neto, Katsnelson, Beenakker, Guinea, Lee, Peres...)
- mobility up to $10^6 \text{ cm}^2/\text{Vs}$; High current density (10^8 - $10^9 \text{ mA}/\mu\text{m}$, or $\mu\text{A}/\text{atomic row}$)

Band Gap Engineering

- What makes graphene such a wonderful conductor also gives rise to its critical weakness
 - Gapless: not useful for digital electronics
- Band gap engineering

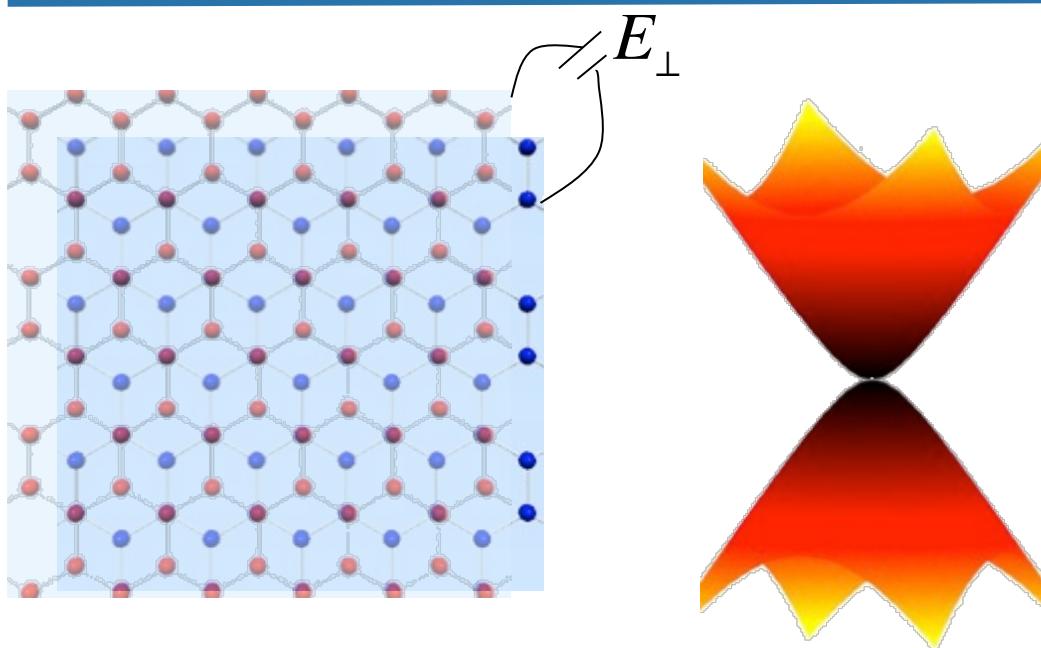


- Many proposals
 - Nanoribbons
 - Chemical modification
 - Strain-based engineering
 - Bilayer and trilayer
- } most cases: transport gap induced by disordered edges,
not true band gap
→ requires very large strain

Bilayer and Trilayer

- ✓ Truly 2D material
- ✓ Quadratic and cubic Dispersion Relations: massive Dirac fermions
- ✓ Surface 2DEG with tunable charge density and type
 - ✓ Optical, STM and mechanical measurements
 - ✓ Easily coupled to special electrodes (superconductors, ferromagnets)
- ✓ the ultimate elastic membrane
 - ✓ Morphology \leftrightarrow electronic properties
- ✓ High Mobility (up to 10^5 – 10^6 cm²/Vs)
- ✓ High thermal conductivity
- ✓ Chemically inert and stable
- ✓ Tunable band gaps (both single-particle and interaction-induced)
- ✓ Interaction-induced collective states

Bilayer Graphene (BLG) – Single Particle Picture

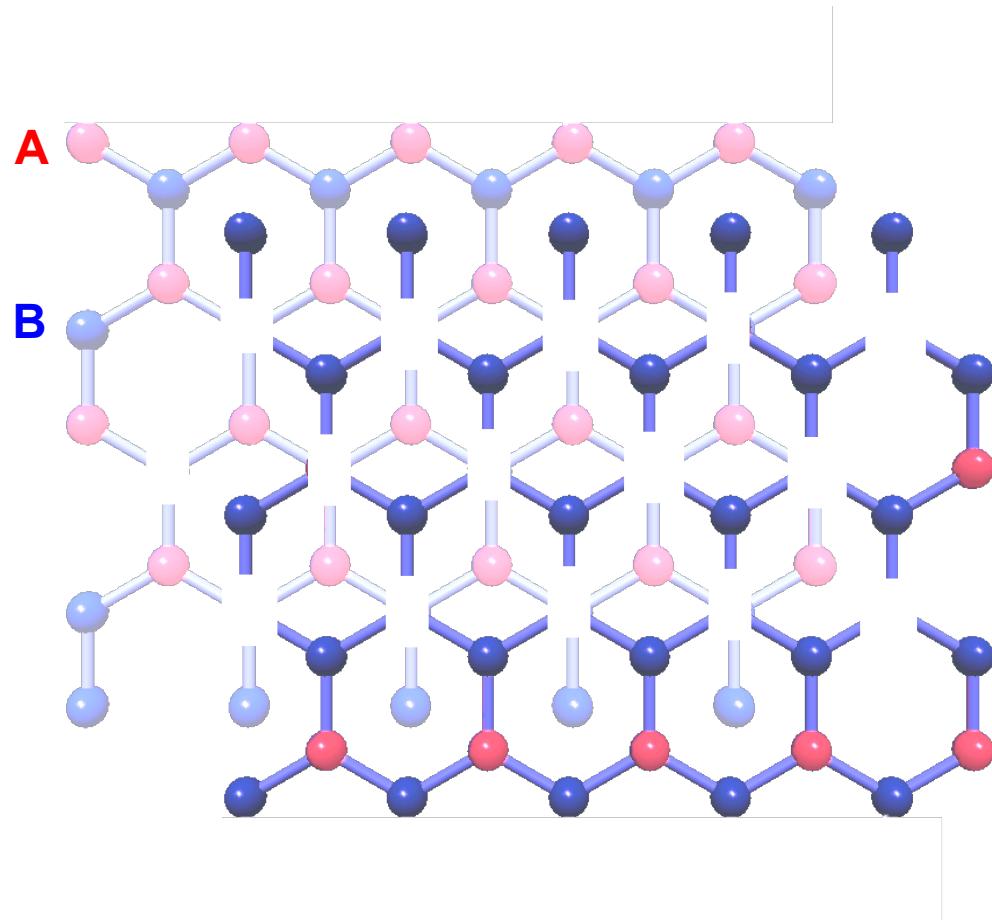


- Quadratic dispersion with zero band gap

McCann PRB 2006; McCann & Fal'ko PRL 2006; Castro *et al* PRL 2007; Castro Neto *et al* PRB

Morpurgo group, Nature Materials 2008; Avouris group, Nano Lett., 2010; Szafranek *et al*, APL 2010; Wang group, Nature 2009; Zhu group, PRB 2010; Fuhrer group, Nano Lett. 2010; Jarillo-Herrero group, PRL 2010; Lau group, Nano Lett. 2010.

Bilayer Graphene – Single particle gap

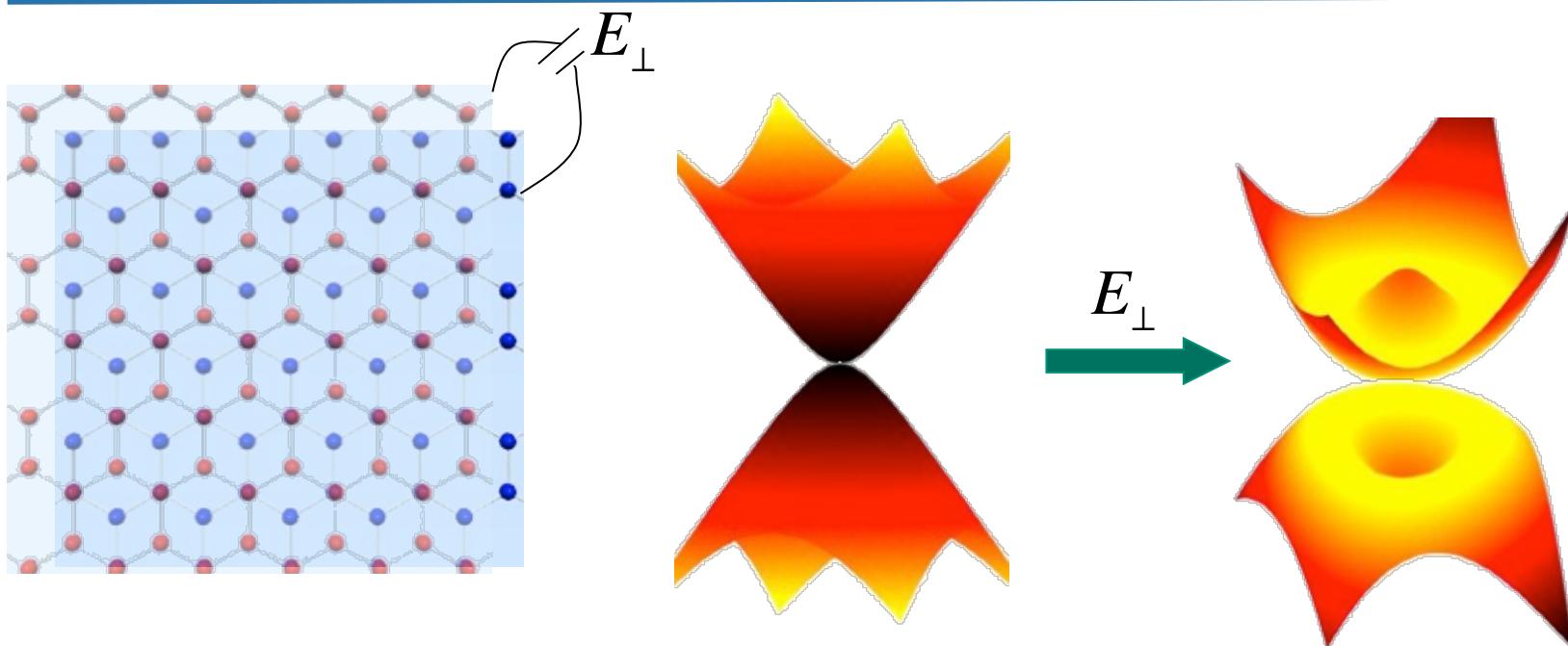


- Stacked atoms hybridize to form higher energy bands
- Left with A sub-lattice from top layer and B sub-lattice from bottom layer
- A-B symmetry \leftrightarrow layer symmetry
- Out-of-plane electric field breaks A-B symmetry \rightarrow generate a gap

Lui et al Nano Lett. 2010

Aoki et al, *Solid state commun.* 2007; McCann PRB 2009; Guinea et al PRB 2006

Bilayer Graphene (BLG) – Single Particle Picture

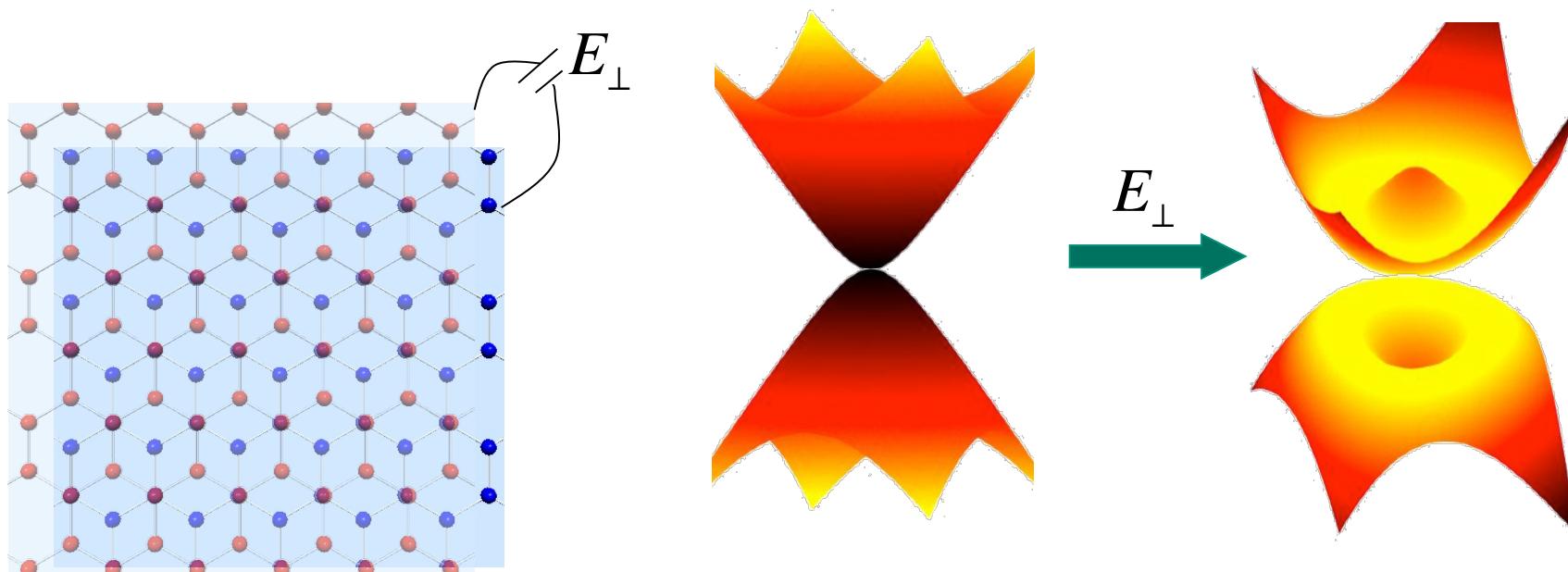


- Quadratic dispersion with zero band gap
- Perpendicular electric field induces band gap

McCann PRB 2006; McCann & Fal'ko PRL 2006; Castro *et al* PRL 2007; Castro Neto *et al* PRB

Morpurgo group, Nature Materials 2008; Avouris group, Nano Lett., 2010; Szafranek *et al*, APL 2010;
Wang group, Nature 2009; Zhu group, PRB 2010; Fuhrer group, Nano Lett. 2010; Jarillo-Herrero group, PRL 2010;
Lau group, Nano Lett. 2010.

Bilayer Graphene (BLG) – Band Structure

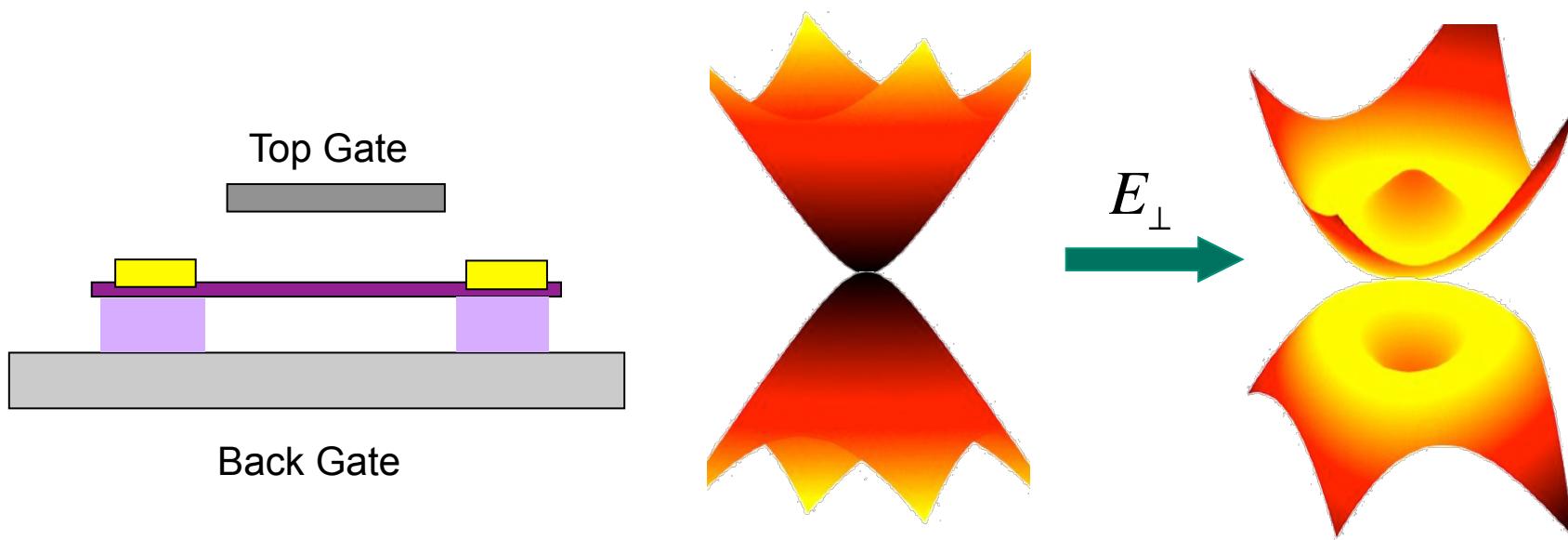


McCann PRB 2006; McCann &
Fal'ko PRL 2006; Castro *et al*
PRL 2007; Castro Neto *et al* PRB

- Quadratic dispersion with zero band gap
- Perpendicular electric field E_{\perp} breaks sublattice and inversion symmetry
→ opens band gap
- Tunable Band gap (\propto electric field)

Morpurgo group, Nature Materials 2008; Avouris group, Nano Lett., 2010; Szafranek *et al*, APL 2010;
Wang group, Nature 2009; Zhu group, PRB 2010; Fuhrer group, Nano Lett. 2010; Jarillo-Herrero group, PRL 2010;
Lau group, Nano Lett. 2010.

Necessity for Dual Gates



- In single-gated devices, E_{\perp} scales with charge density n
- Dual-gated devices:

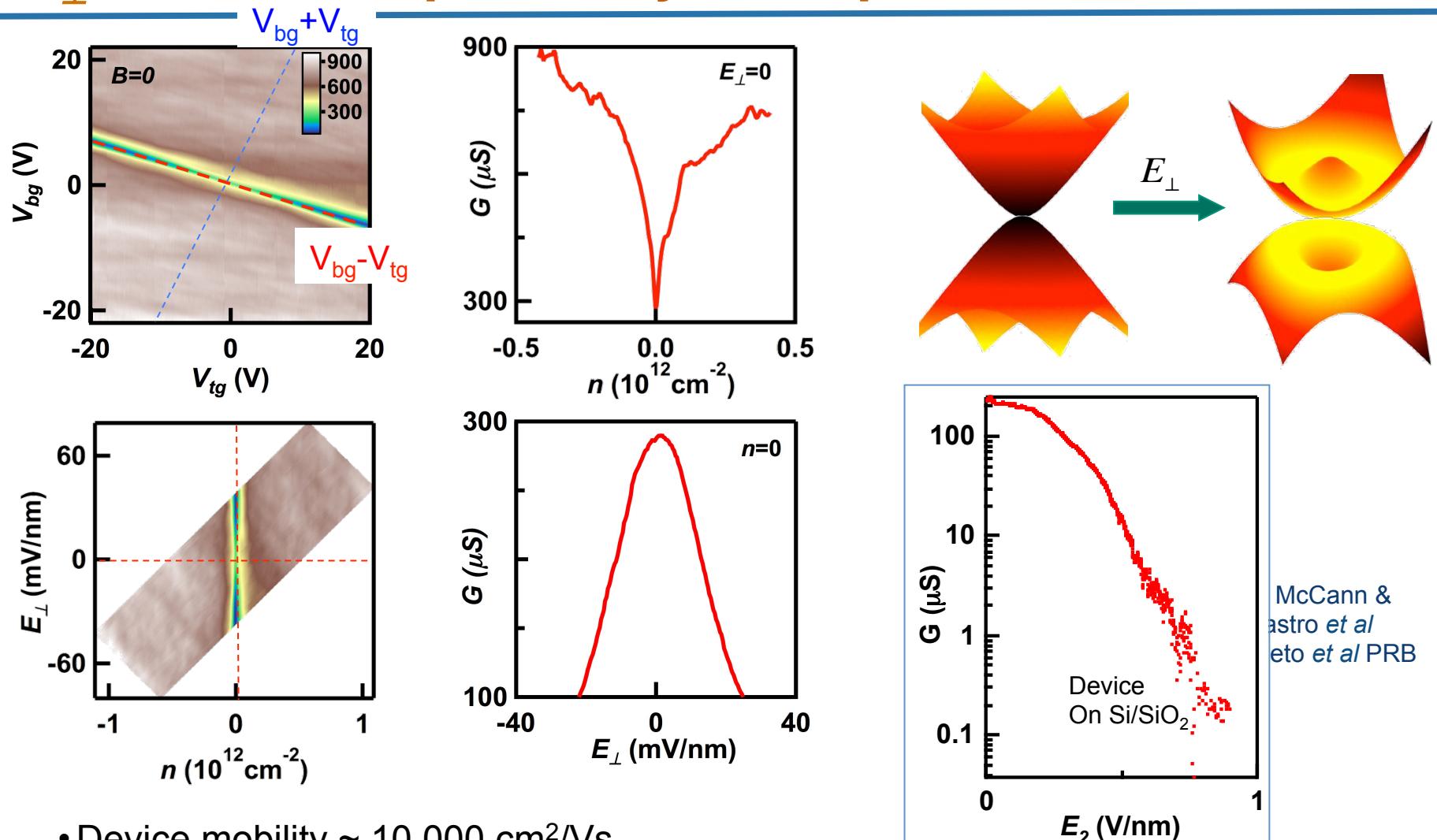
$$n \propto V_{bg} + V_{tg}$$

$$E_{\perp} \propto V_{bg} - V_{tg}$$

McCann PRB 2006; McCann &
Fal'ko PRL 2006; Castro *et al*
PRL 2007; Castro Neto *et al* PRB

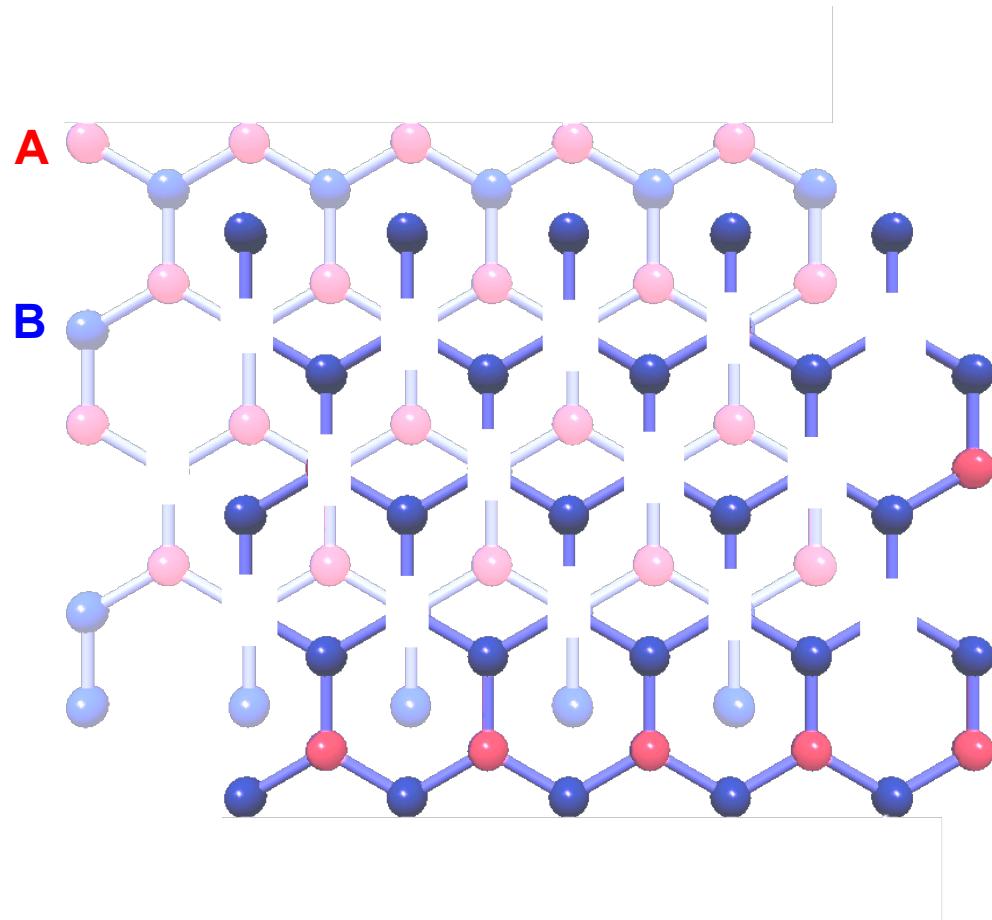
Lau group, APL 2007, New J. Phys. 2008; Savchenko group, Nano Lett. 2007; Marcus group, Science 2007;
Goldhaber-Gordon group, PRL 2007.

E_{\perp} -induced Gap of Bilayer Graphene



- Device mobility $\sim 10,000$ cm²/Vs
- Tunable Band gap (\propto electric field), $G \sim \exp(-E_{\perp})$
- *needs very large voltages*

Bilayer Graphene – Interaction-induced gap

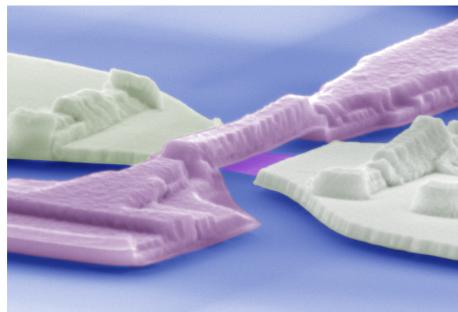


- Stacked atoms hybridize to form higher energy bands
- Left with A sub-lattice from top layer and B sub-lattice from bottom layer
- A-B symmetry \leftrightarrow layer symmetry
electronic interactions spontaneously
- ~~Out-of-plane electric field~~ breaks A-B symmetry → generate a gap

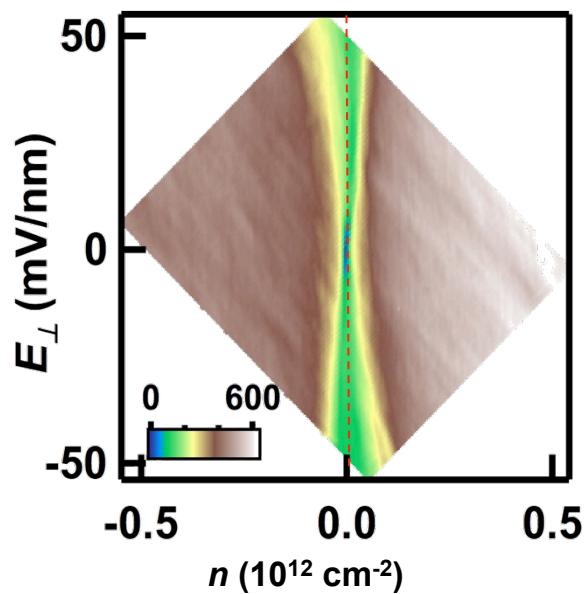
Lui et al Nano Lett. 2010

Aoki et al, Solid state commun. 2007; McCann PRB 2009; Guinea et al PRB 2006

High Mobility Bilayer Devices

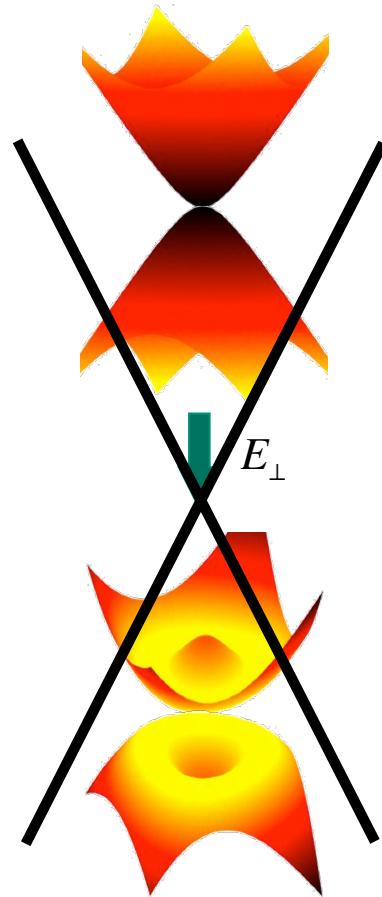
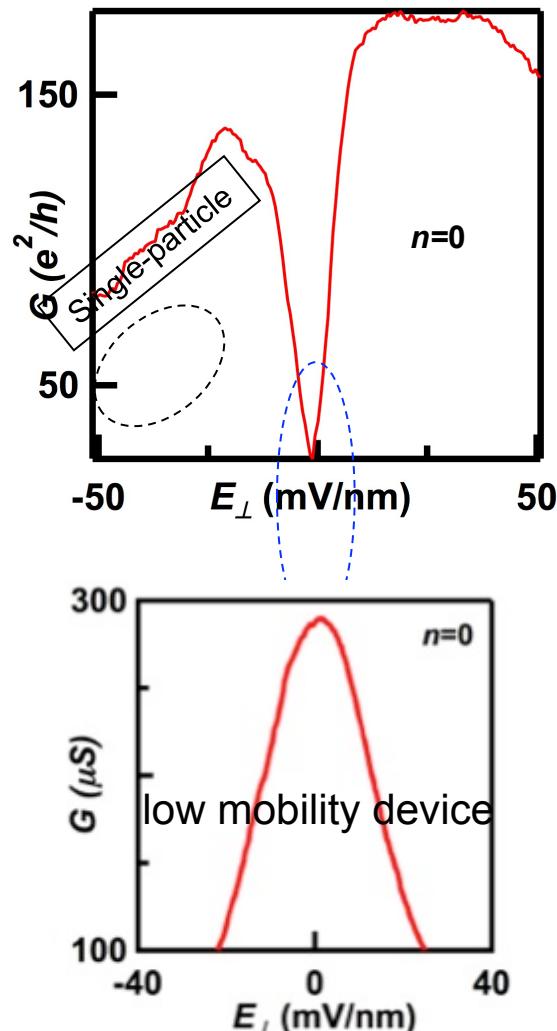


Device mobility $\sim 80,000 \text{ cm}^2/\text{Vs}$



Lowest conductance (insulating) at $n=E=0$.

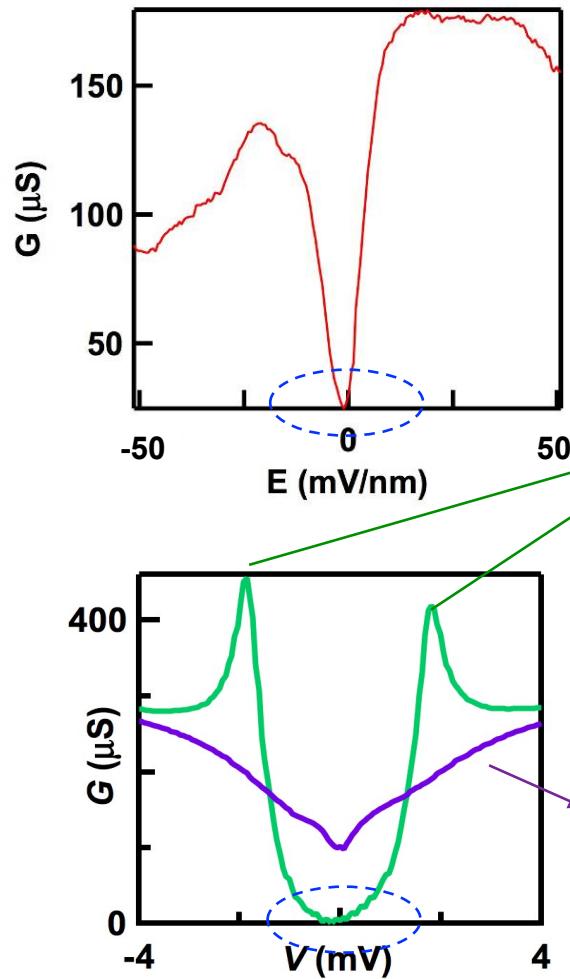
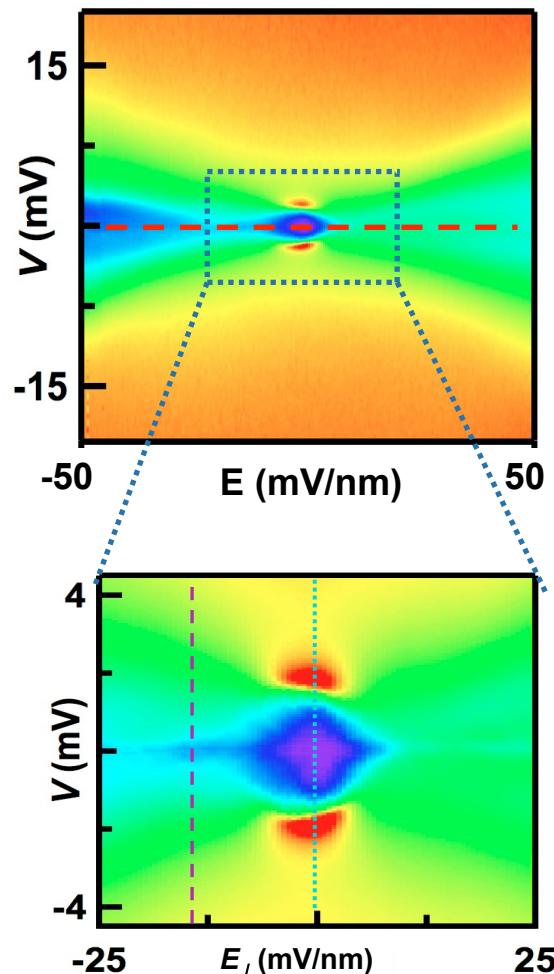
- contradicts single particle picture



See also R. T. Weitz, M. T. Allen, B. E. Feldman, J. Martin, A. Yacoby, Science, 330, 812 (2010).

Insulating State and Intrinsic Gap

dI/dV vs. Electric field and source-drain bias at charge neutrality point



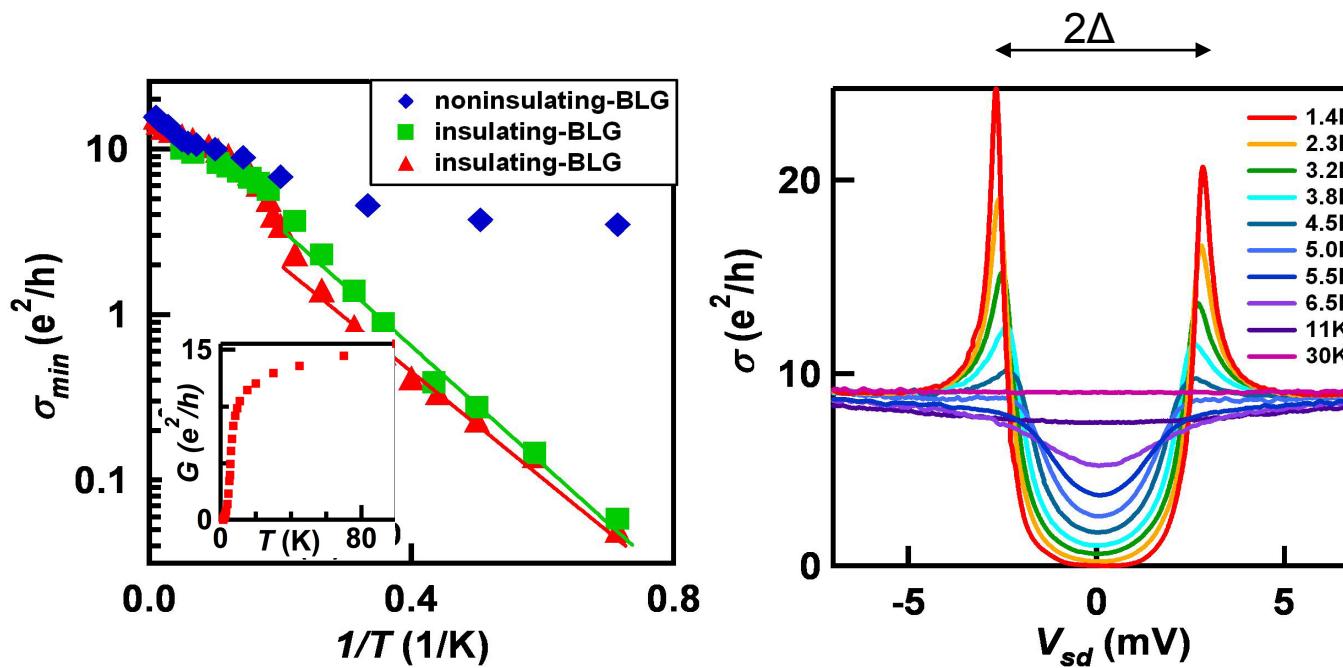
Insulating at $V=n=E=0$
($R \sim M\Omega$)

dI/dV resembles the density of states of a gapped state
→ an **intrinsic gapped** ground state $\sim 2\text{meV}$

- Gap can be closed by electric field of either polarity $\sim 12\text{ meV/nm}$.

J. Velasco Jr., L. Jing, W. Bao, Y. Lee, P. Kratz, V. Aji, M. Bockrath, C.N. Lau, C. Varma, R. Stillwell, D. Smirnov, Fan Zhang, J. Jung, A.H. MacDonald, Nature Nanotechnol., 7, 156 (2012).

Temperature Dependence



- Consistent $\sigma_{min}(T)$ dependence among different devices for $5 < T < 130$ K
 - At 5K
 - abrupt change in slope of $\sigma_{min}(T)$
 - deviation between insulating and non-insulating devices
 - dI/dV side peaks disappear
- Transition from insulating to conductive states,
 $T_c \sim 5$ K, activation gap ~ 1.8 meV

W. Bao, J. Velasco Jr, F. Zhang, L. Jing, B. Standley, D. Smirnov, M. Bockrath, A. MacDonald, C.N. Lau, PNAS, 109, 10802 (2012).

Collective State(s) at the Dirac Point in BLG

- Strong interaction at the Dirac point → broken symmetry states

Experimental Work

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An analogy...

Singe Particle Picture



8 million strangers...

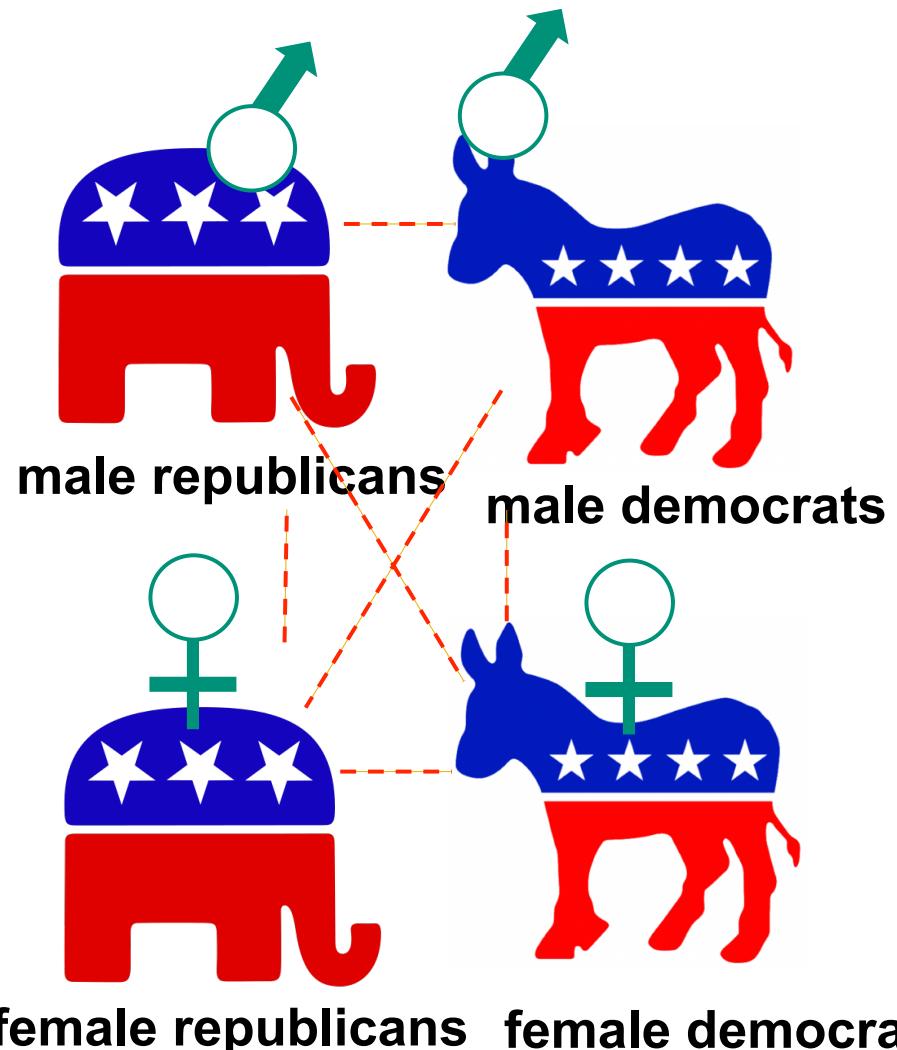
Electronic Interactions



dinner party with assigned seating

→ correlated phases such as
superconductors, magnets,
Mott insulators, etc...

An analogy...



Electronic Interactions



dinner party with assigned seating

Electrons can

- have spin up or down
- K and K' valleys

Collective States at the Dirac Point in BLG

- Strong interaction at the Dirac point → broken symmetry states
- Nature of the states under intense efforts.

Popular Candidates

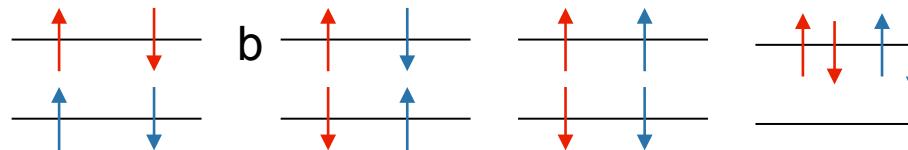
Protected edge states **Bulk gap**

	Nematic Order	Anomalous Hall	Quantum Spin Hall	Layer Antiferromagnet	QVH
Gapped?	No	Yes	Yes	Yes	Yes
2-terminal σ_{min}	finite	$4e^2/h$	$4e^2/h$	0	0
Symmetry Broken	rotation	time reversal; Ising Valley	spin rotational; Ising Valley	time reversal; spin rotation	inversion

red: K electrons
blue: K' electrons

Possible Collective States

	Nematic Order	Anomalous Hall	Quantum Spin Hall	Layer Antiferromagnet	QVH
Gapped?	No	Yes	Yes	Yes	Yes
σ_{min}	finite	$4e^2/h$	$4e^2/h$	0	0
Symmetry Broken	rotation	time reversal; Ising Valley	spin rotational; Ising Valley	time reversal; spin rotation	inversion

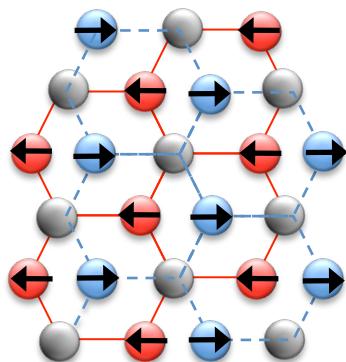


red: K electrons
blue: K' electrons

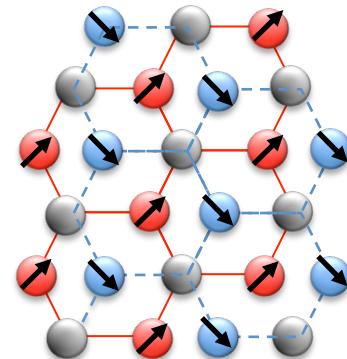
- Electronic interactions gives rise to a intrinsic **gapped, insulating collective state** in charge neutral graphene
- Gap can be closed by electric field of either polarity

Most likely candidate

B=0
Antiferromagnet



B \neq 0
Canted
Antiferromagnet



Layer Antiferromagnet
Gapped
Insulating
Breaks time reversal & spin rotation symmetry

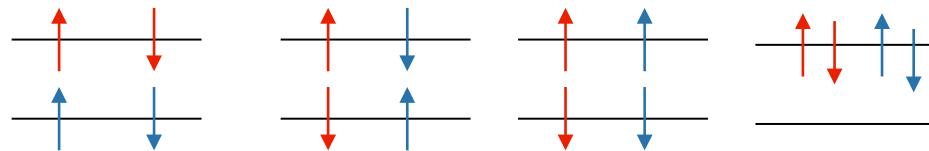
- Electronic interactions gives rise to a intrinsic **gapped, insulating collective state** in charge neutral graphene
- Gap can be closed by electric field of either polarity

Collective States

Protected edge states
~ topological insulators

Bulk
gap

	Nematic Order	Anomalous Hall	Quantum Spin Hall	Layer Antiferromagnet	QVH
Gapped?	No	Yes	Yes	Yes	Yes
σ_{min}	finite	$4e^2/h$	$4e^2/h$	0	0
Symmetry Broken	rotation	time reversal; Ising Valley	spin rotational; Ising Valley	time reversal; spin rotation	inversion



red: K electrons
blue: K' electrons

Outstanding Questions

- Is the insulating state antiferromagnetic?
- Is it the ground state?
- Can we engineer BLG to other ordered, collective states, e.g. with protected edge states, dissipationless transport? (tentative yes)

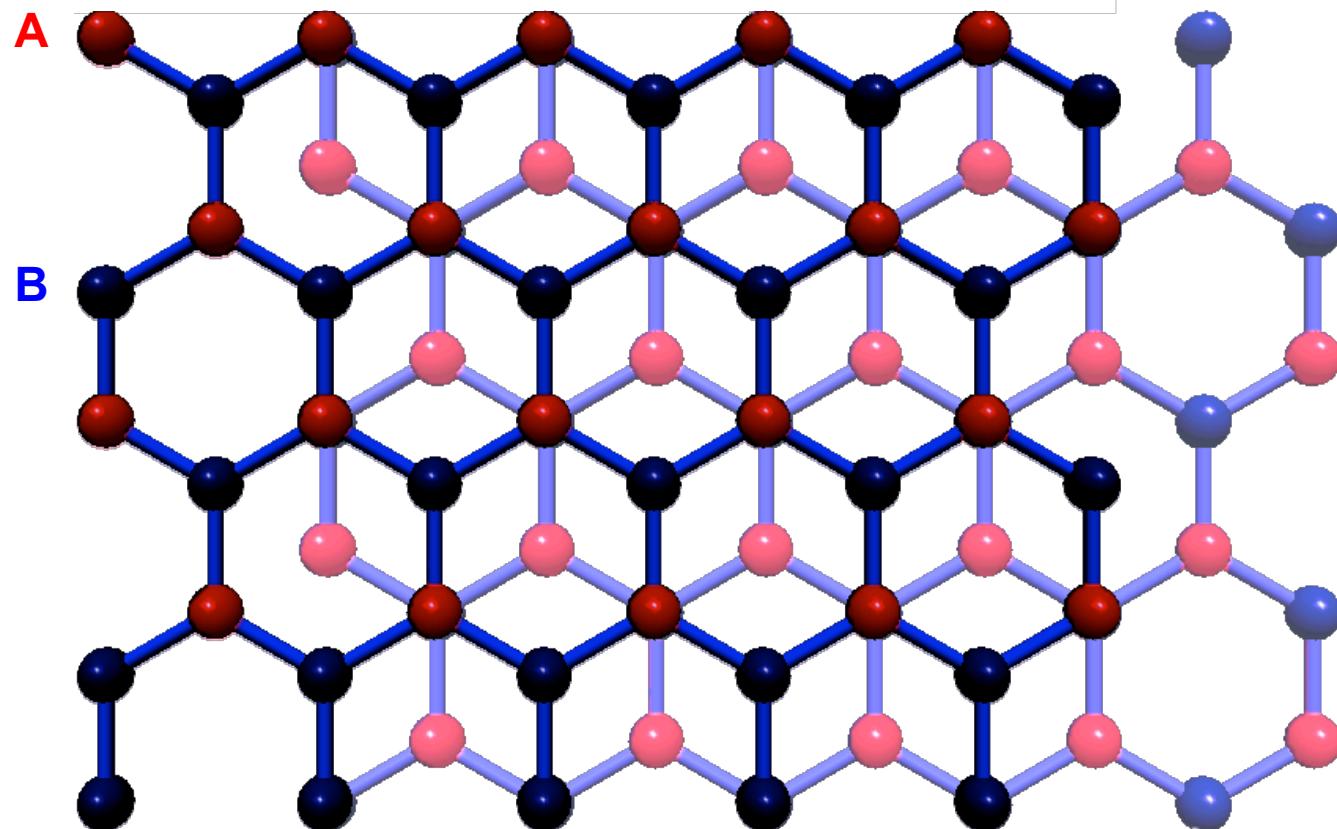
Conclusion

- Insulating state with an intrinsic gap $\sim 2\text{-}3 \text{ meV}$ in bilayer graphene
- Gap can be tuned by temperature, disorder, electric field and density
- Can we realize transition among various collective states?
- Source-drain bias spectroscopy to resolve symmetry-broken Landau level gaps, which are strongly electric field dependent

Why stop at 2?

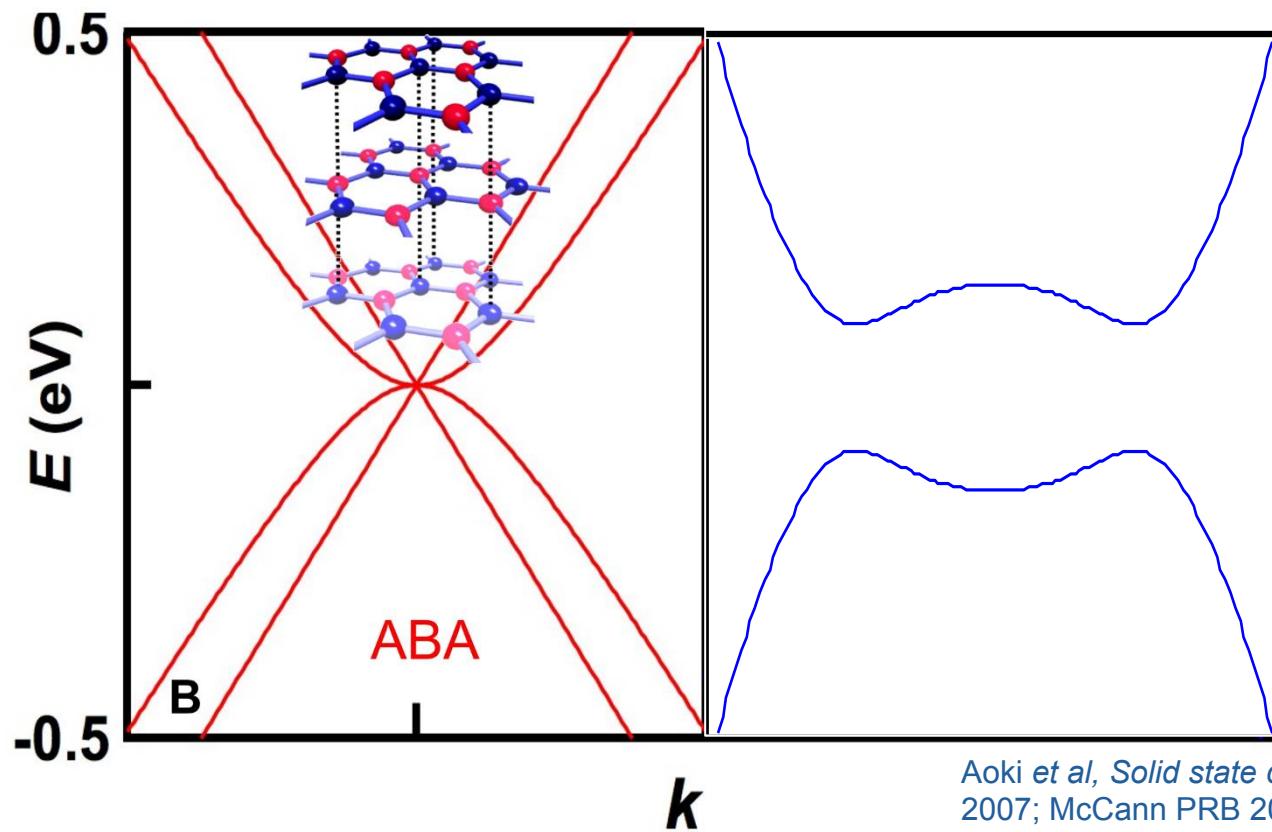
Trilayer Graphene

- ABA (Bernal) stacking
- ABC (rhombohedral) stacking
- can be distinguished using Raman spectroscopy



Aoki et al, Solid state commun. 2007; McCann PRB 2009; Guinea et al PRB 2006

Trilayer graphene – Single Particle Picture



Aoki *et al*, *Solid state commun.*
2007; McCann PRB 2009; Guinea *et
al* PRB 2006

$$E_{\perp} = 0$$

$E \sim$ monolayer band
+ bilayer band

$$E \sim \hbar^3 k^3$$

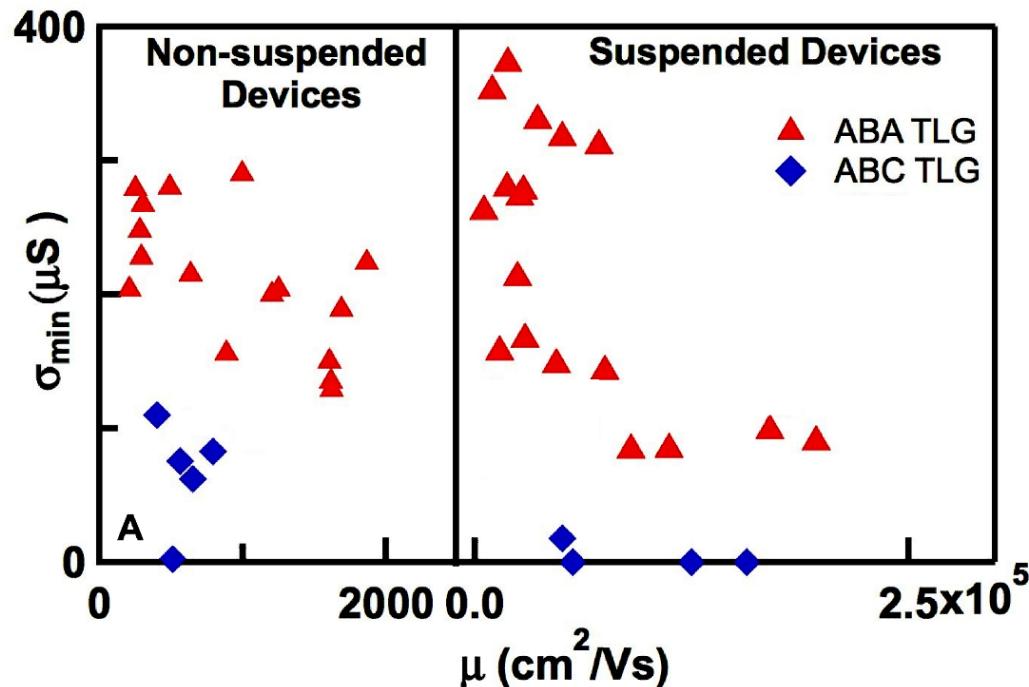
$$E_{\perp} \neq 0$$

Tunable Band Overlap

Tunable Band Gap

Craciun *et al*, *Nature Nanotech.* 2009; Lui *et al* *Nature Phys.* 2011; Bao *et al* *Nature Phys.* 2011, Taychatanapat
et al *Nature Phys.* 2011; Henriksen *et al* *PRX* 2012; Zhang *et al* *Nature Phys.* 2011; Zhu group *Nano Lett.* 2013.

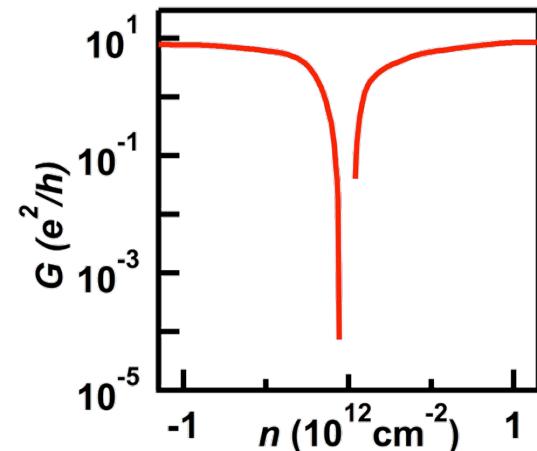
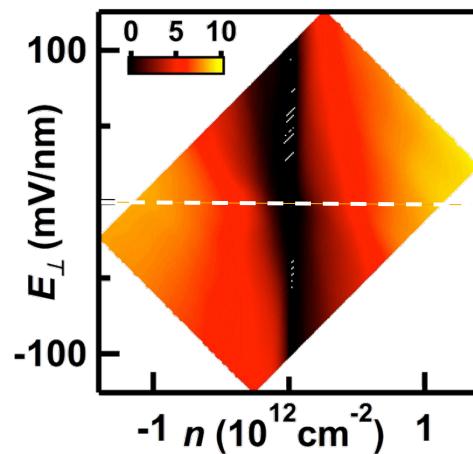
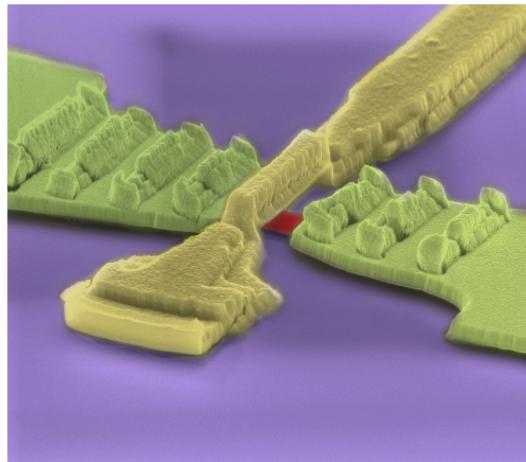
Minimum conductivity (σ_{min})



- Stacking orders identified by Raman spectroscopy
- σ_{min} ABA-stacked trilayer remains finite $>100 \mu\text{S}$.
- σ_{min} of ABC-stacked trilayer is significantly smaller, ~ 0 for suspended devices.

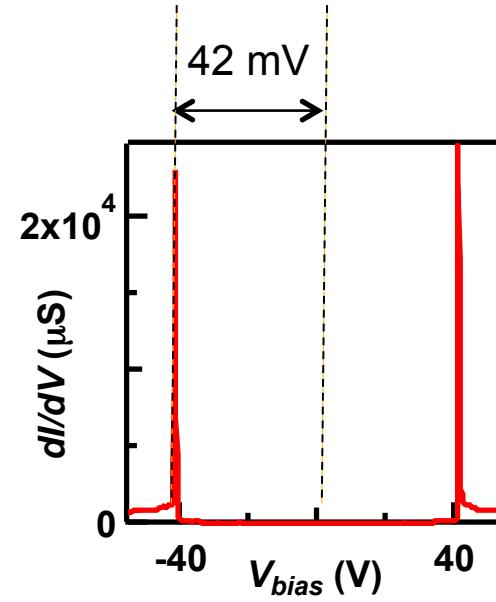
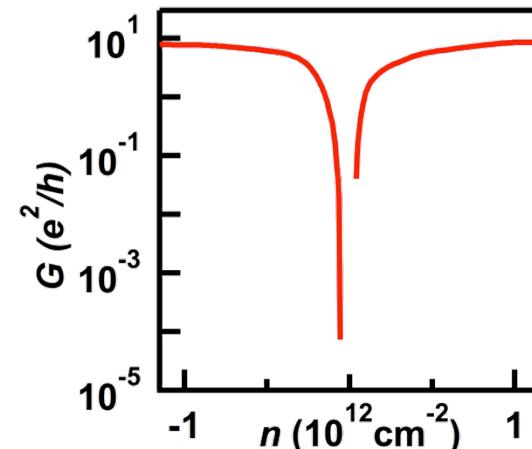
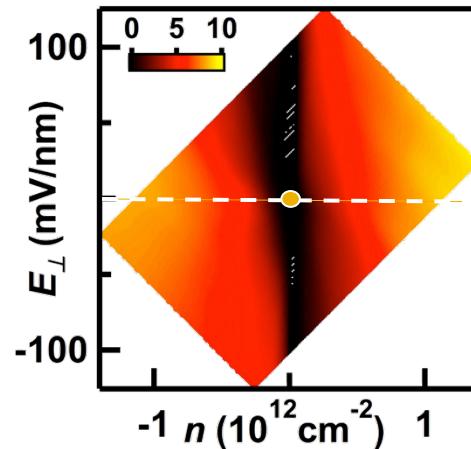
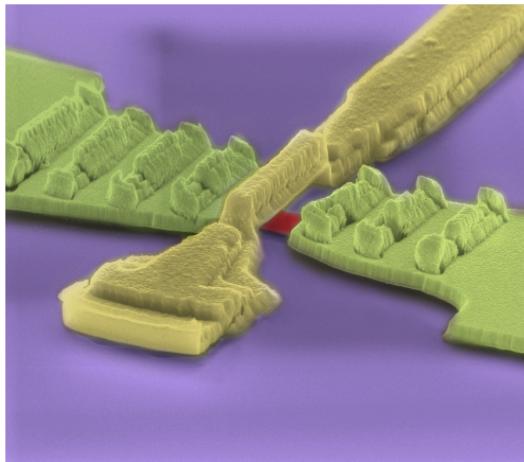
W. Bao, L. Jing, J. Velasco Jr., Y. Lee, G. Liu, D. Tran, B. Standley, M. Aykol, S. B. Cronin, D. Smirnov, M. Koshino, E. McCann, M. Bockrath, and C.N. Lau, Nature Phys. (2011)

Dual-Gated Suspended ABC Trilayer Devices



- mobility 20,000 – 90,000 cm²/Vs
- Device is insulating at $n = E_{\perp} = 0$
- Insulating state persists for large range of E_{\perp}
- Failure of single particle picture

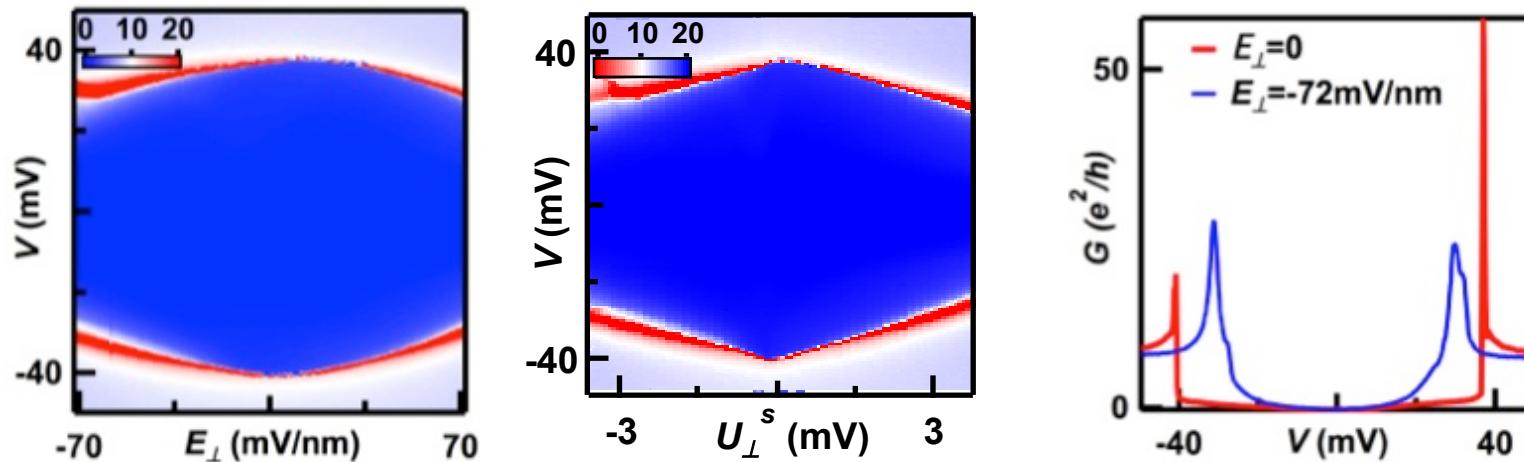
Intrinsic Insulating state



- mobility 20,000 – 90,000 cm^2/Vs
- Device is insulating at $n = E_\perp = 0$
- Insulating state persists for large range of E_\perp
- Failure of single particle picture
- $\Delta \sim 42 \text{ meV}$

Effect of electric field

Differential conductance G vs source drain bias V at $n=0$



Lee et al, submitted.

- intrinsically insulating at $n=E_{\perp}=0$, a gap ~ 42 meV
- gap closes symmetrically with applied E_{\perp} (linearly after corrected for screening)
- gap not completely closed at largest $|E_{\perp}|$
 - not a single particle gap
 - arises from electronic interactions

Recent theories: MacDonald, Vafek, Xie, Honerkamp, Vozmediano, Barlas

Interaction Parameter

$$\alpha \sim \frac{\text{Coulomb Energy}}{\text{Fermi Energy} \sim k^p}$$

$$\sim \kappa^{-1} n^{-\frac{p-1}{2}}$$

n =charge density (10^{10} cm^{-2})
 κ =dielectric constant

	Dispersion	$\alpha (\kappa=1)$	Interaction-induced Gap
GaAs/AlGaAs	$E \sim k^2$	$(10-50)/\sqrt{n}$	
Single Layer Graphene	$E \sim k$	2.2	<0.2 meV
Bilayer Graphene	$E \sim k^2$	$70/\sqrt{n}$	2-3 meV
ABC-stacked Trilayer	$E \sim k^3$	$1500/n$	42 meV
ABC-stacked N-layer	$E \sim k^N$	gigantic	300 meV?

Interaction-induced gap in tetra-layer?

Conclusion



Challenges and Opportunities

- Large scale growth with controlled stacking order
- Controlling impurities and substrate