Massachusetts Institute of Technology

Remote epitaxy through graphene for producing wafer-scale freestanding 3D and 2D materials

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Mechanical Engineering
Materials Science and Engineering
Research Laboratory of Electronics
Microsystem Technology Laboratories

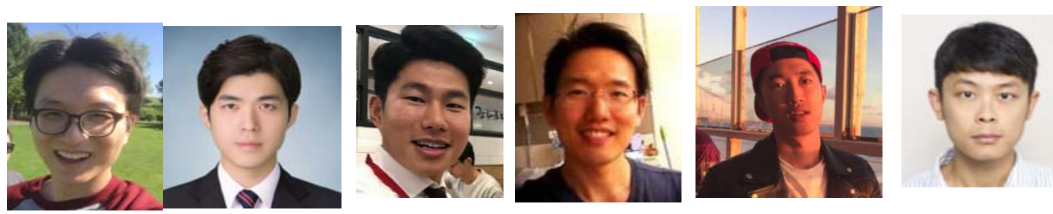
Nanoelectronics Group at MIT

Remote Epitaxy Group

III-V devices *Wide band gap devices* *Functional oxide devices*

Wafer-scale 2D Materials Group

Wearable Electronics /heterointegration Group



Neuromorphic Computing Group

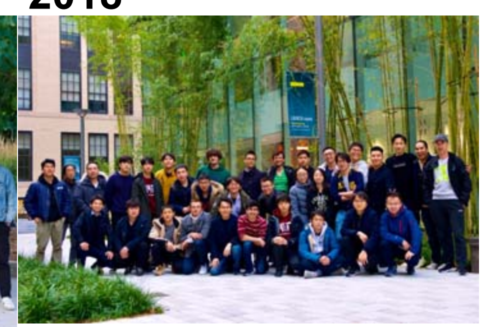
Materials/devices *ANN Arrays* *Algorithm/Training*

Oct 2015

2016

2017

2018



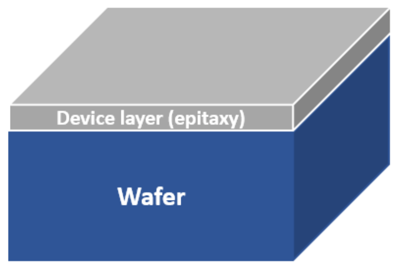
Massachusetts Institute of Technology

Wafer-scale freestanding 3D materials and their heterostructure

Jeehwan Kim
Research Group
http://jeehwanlab.mit.edu

Major challenges: Lattice mismatch + Wafer price

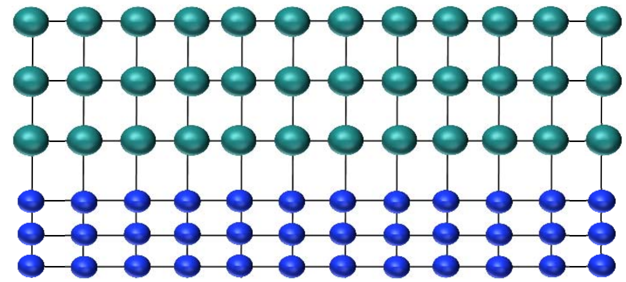
Epitaxy Copy of crystalline information of wafer→ forming device layers



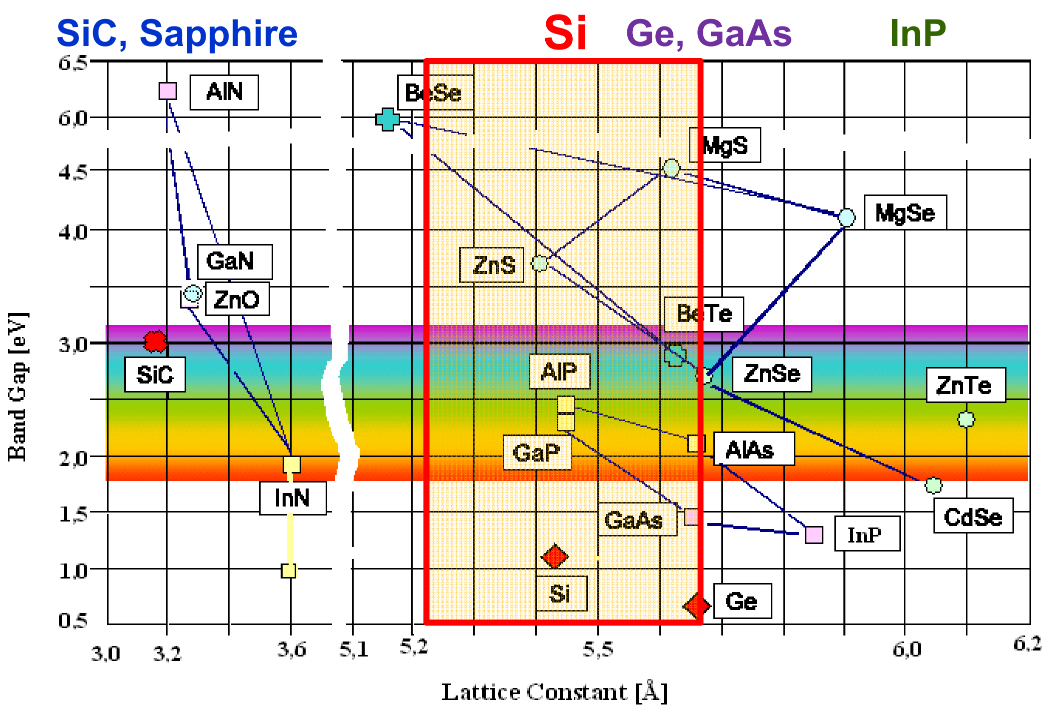
Device Active Region:
~ 100's nm – 1's µm

Substrate thickness:
~ 1 mm

< ~ 1% per volume
(recall how expensive substrate was)



Epitaxy copies crystalline information of the substrate

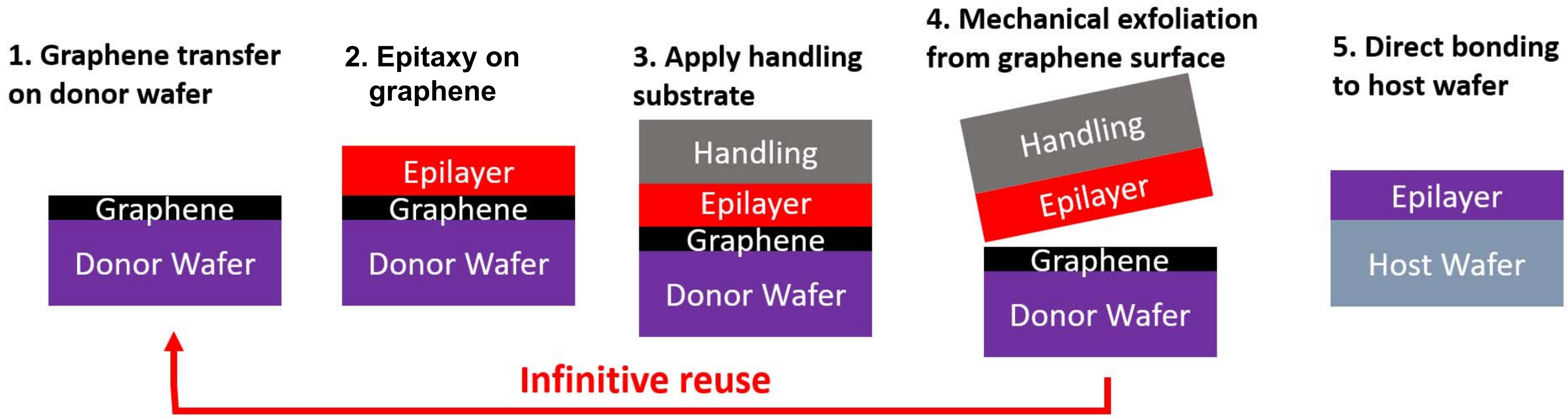


Wafer Price:
SiC > InP > GaAs > Ge >>> Si

Extremely high wafer cost prohibits advancement of current electronics (Non-Si electronics)

Wafer recycling via layer transfer

2D material based layer transfer (2DLT)



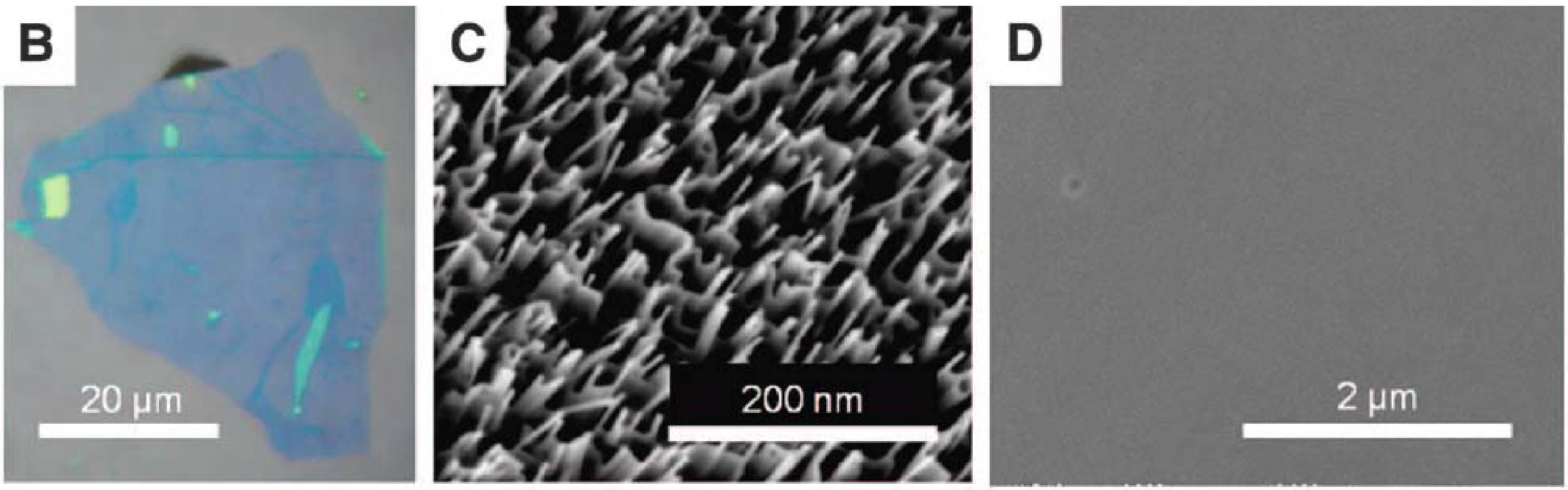
- **sp^2-bonded graphene: No broken bonds on the surface**
  - Precise release from graphene
  - Post-release treatment NOT required
  - **1 sec** release due to weak interaction
  - **Universal for any materials**

	Reusability	Need for post-release treatment	Release rate	Control of release thickness	Universality
Chemical	Mid	Yes	Very slow	Excellent	Low
Optical	Mid	Yes	Slow	Excellent	Low
Mechanical	Low	Yes	Fast	Good	High
Smart-cut	Mid	Yes	Fast	Good	Low
2DLT	**High**	**No**	**Fast**	**Excellent**	**High**

First planar layer transfer process using graphene

Prof. Yi's lab in SNU

K. Chung, C.-H. Lee, and G.-C. Yi, Science, 330, 655–657 (2010).

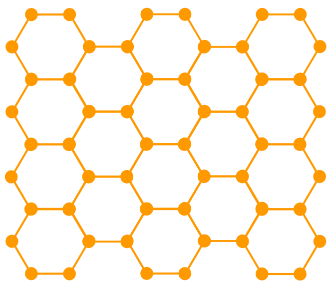


ZnO nanowall wetting layer

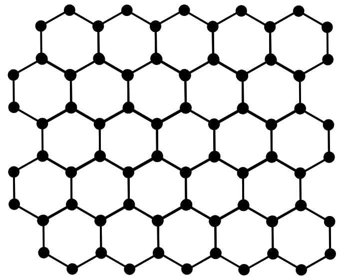
First perfect single-crystalline GaN on graphene (IBM)

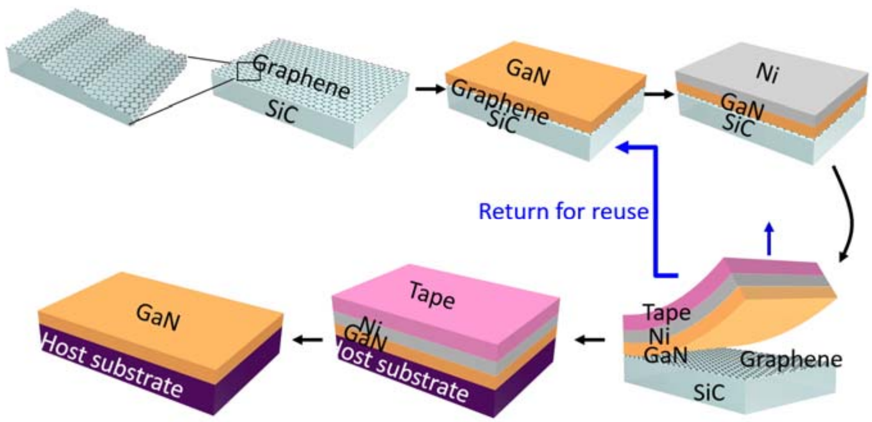
Growth/transfer Single-crystalline GaN on graphene

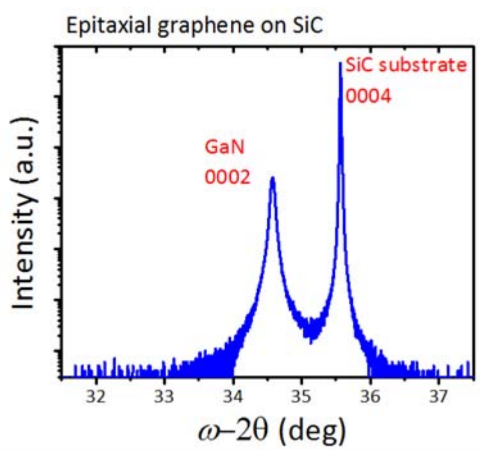
GaN lattice (top view)



Graphene lattice (top view)

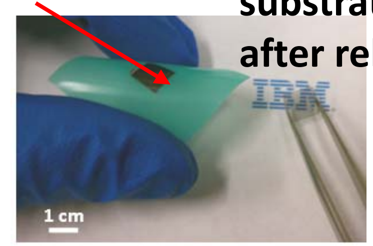


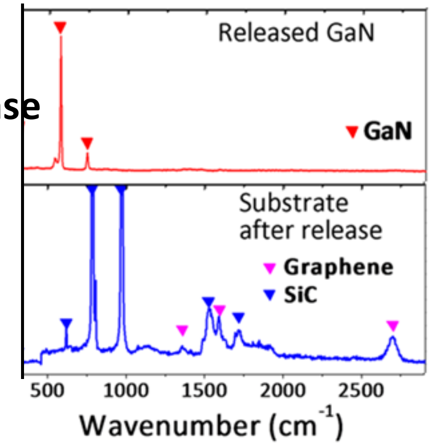




Single-crystalline GaN is perfectly grown

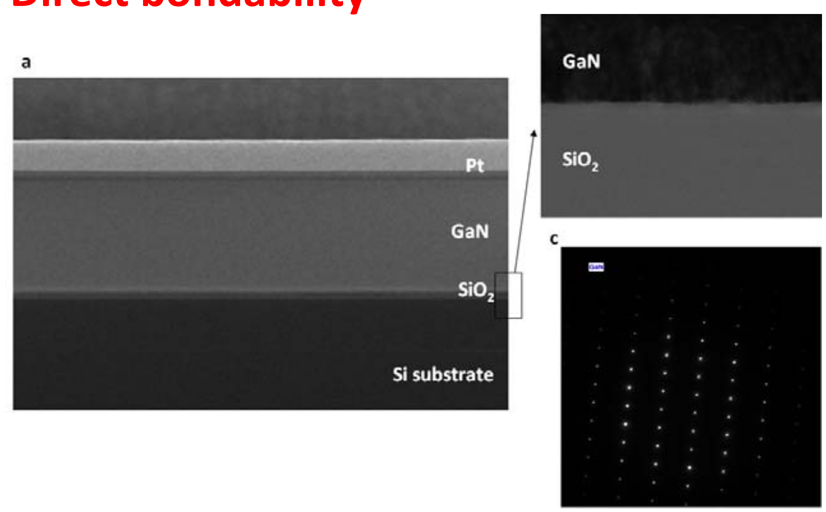
Substrate reusability





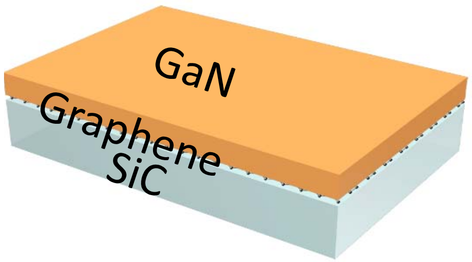
Graphene exists on wafer after GaN peeling
→ No post-treatment required for further recycle

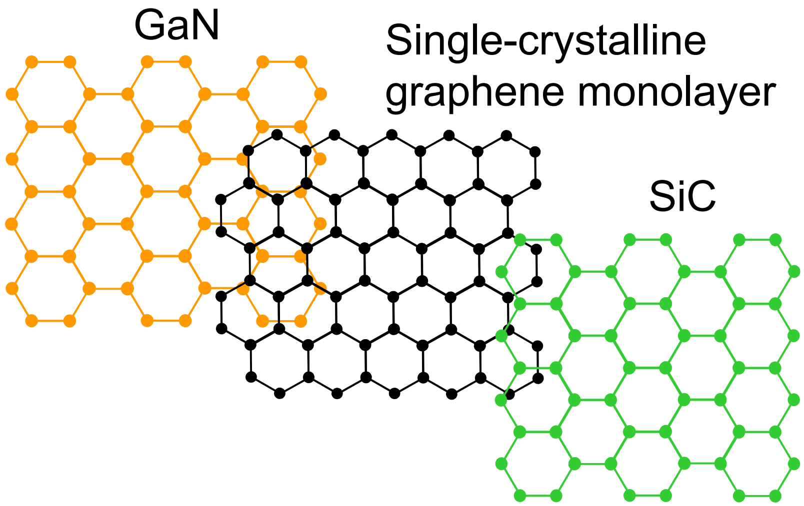
Direct bondability



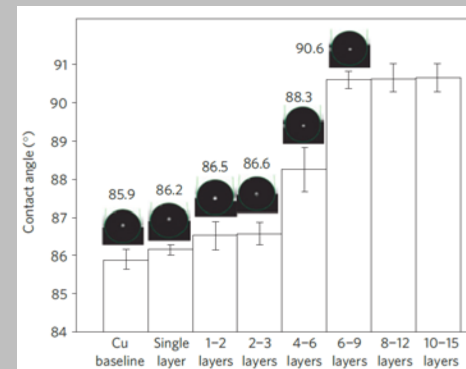
J. Kim et al., ***Nature Communications***, Vol. 5, 4836 (2014)

Where is the seed layer? Graphene or SiC?



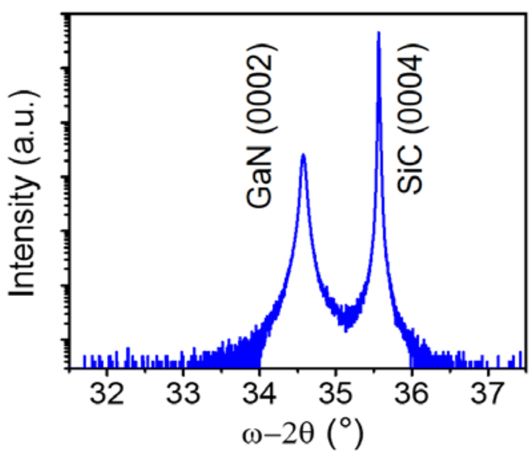


Nature Materials, 11, 217 (2012)



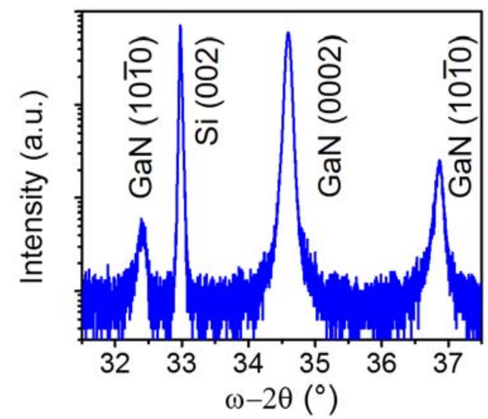
Similar to
wetting transparency?

GaN
on single-crystalline graphene
on single-crystalline SiC



Single-crystalline GaN

GaN
on single-crystalline graphene
on amorphous oxide

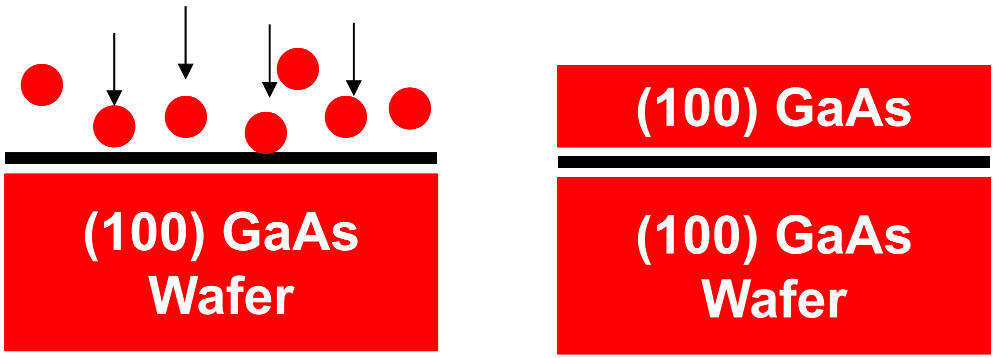


Polycrystalline GaN

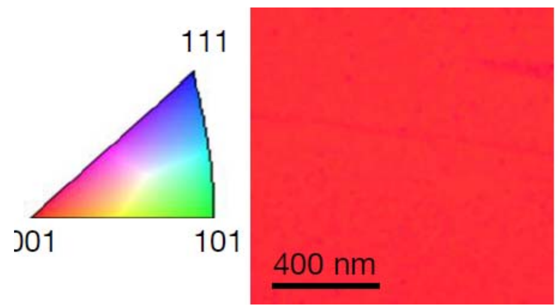
“Remote epitaxy” invented!

Monolayer graphene transparency for epitaxial growth
- Remote homoepitaxy

GaAs epitaxy on graphene monolayer

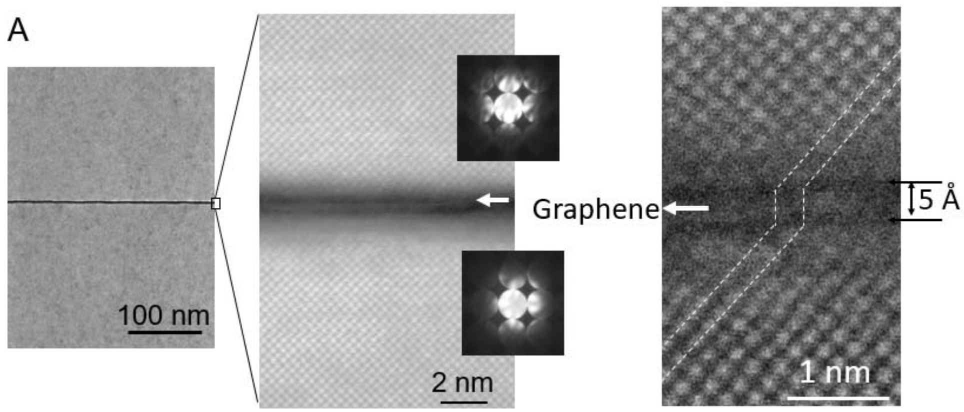


Map of
electron backscatter diffraction (EBSD)

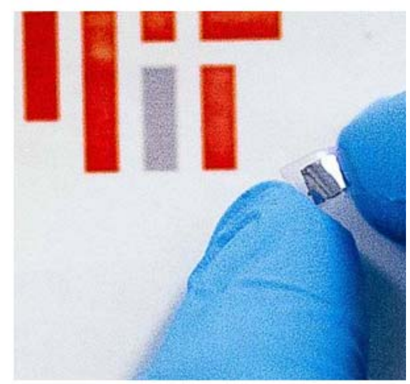


Perfect single-crystalline GaAs
by mimicking substrate

High resolution transmission electron microscopy



Easy exfoliation of GaAs
from graphene surface

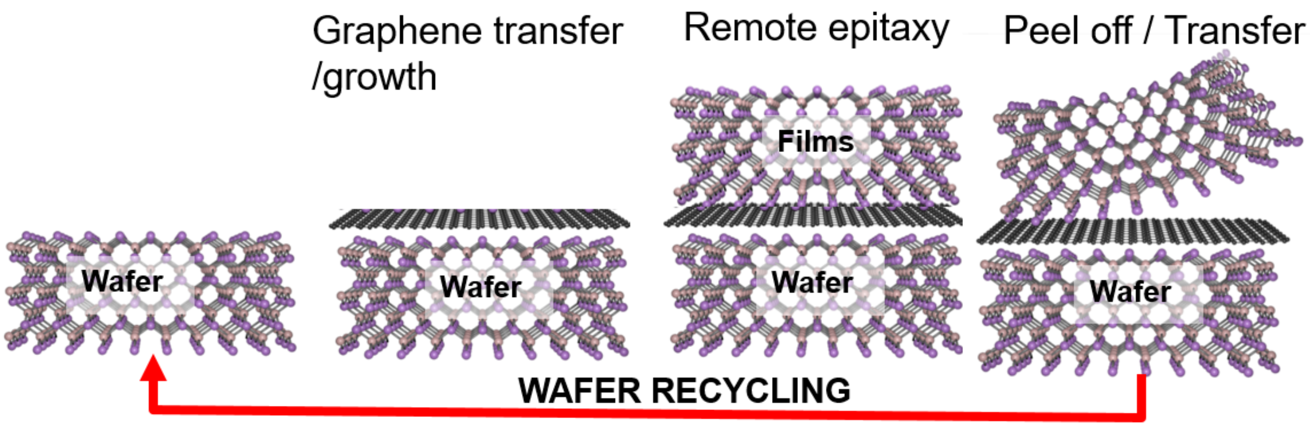


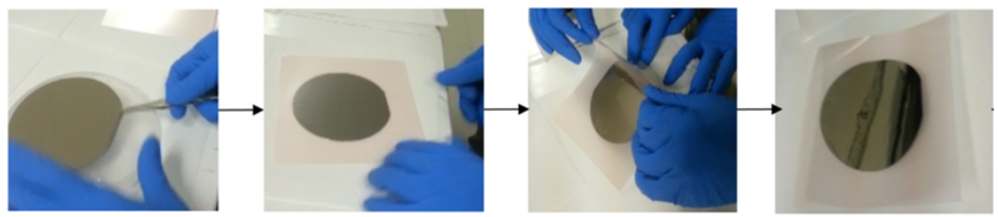
In collaboration with Eugene Fitzgerlad

Y. Kim et al., and J. Kim., ***Nature*** 544, 340 (2017)

2D material-based layer transfer via Remote epitaxy

Application: 2D material-based layer transfer (2DLT)



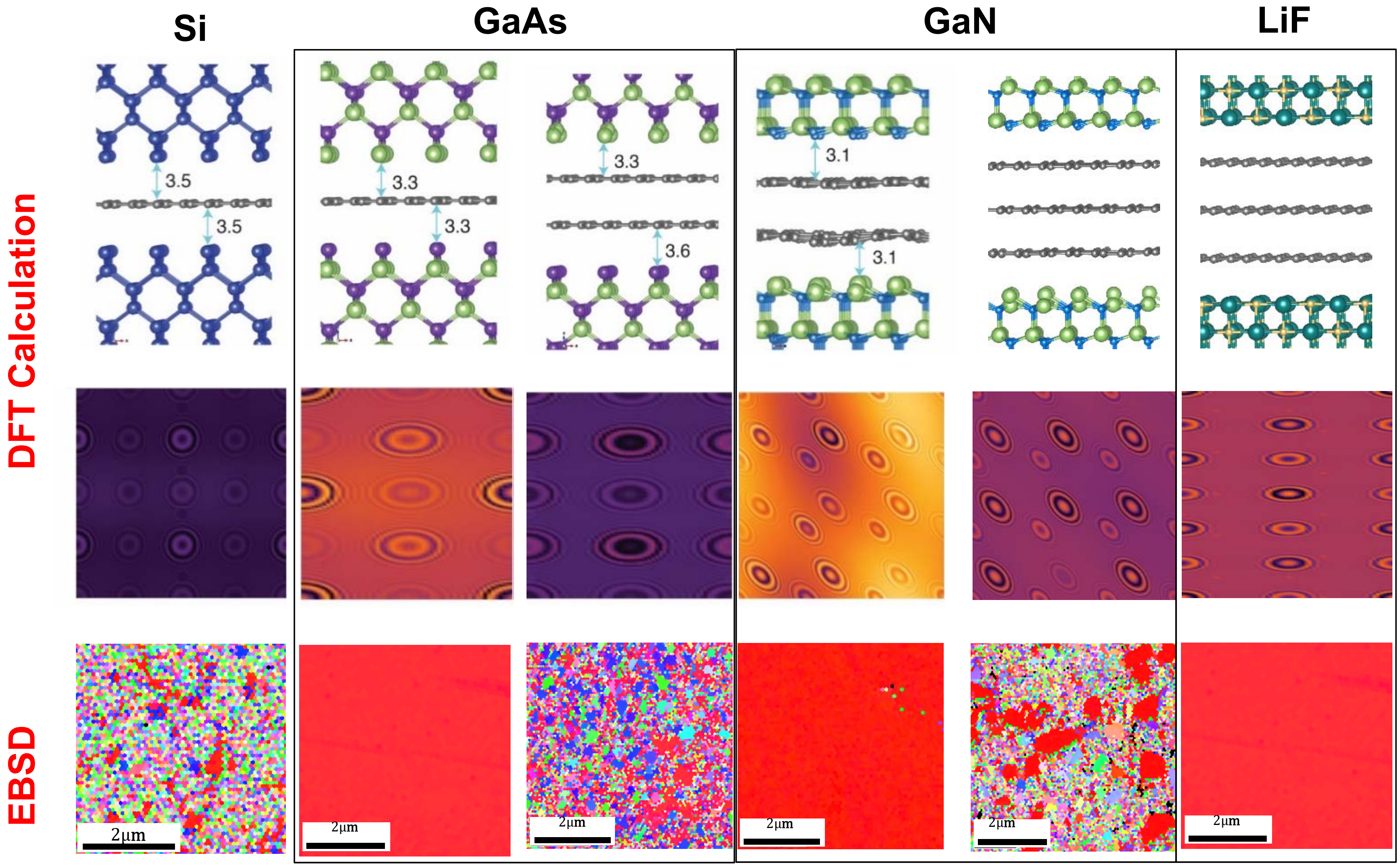


Monolayer graphene + Wafer

= Copy Machine (Film producer)

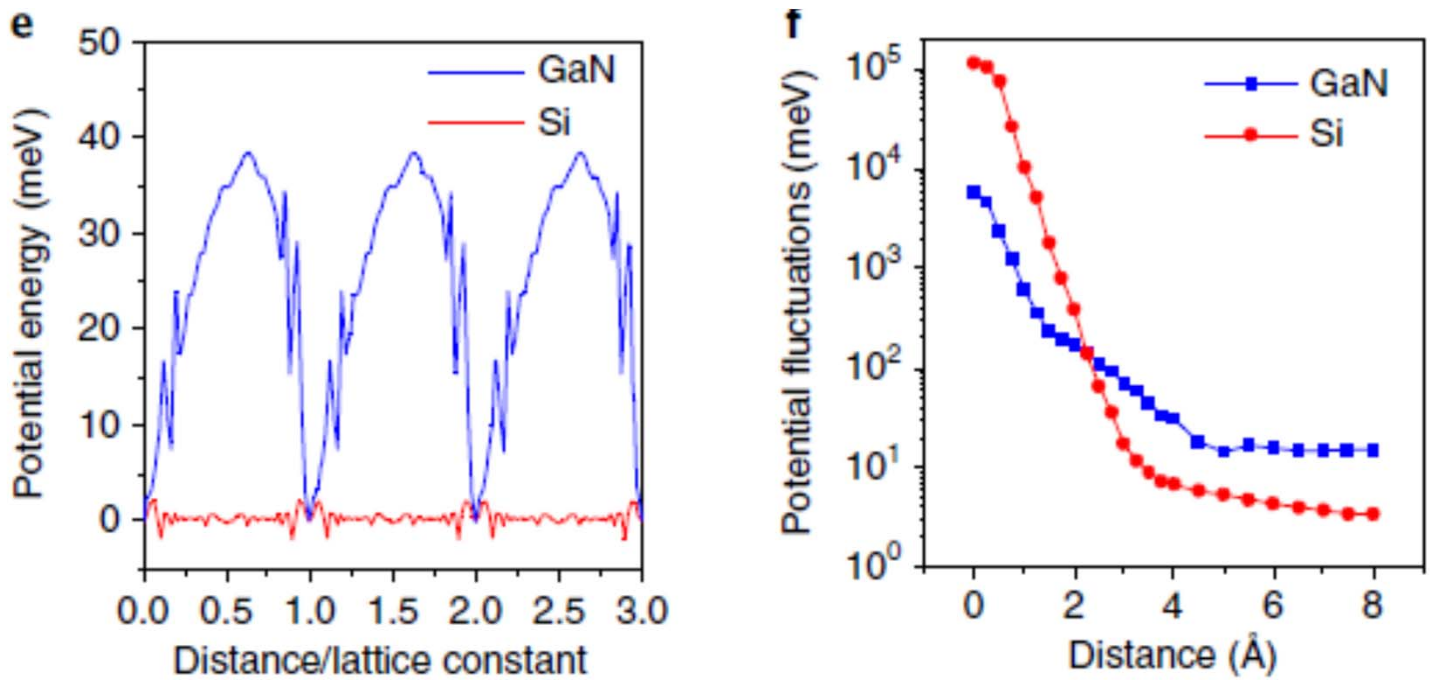
Y. Kim et al., and J. Kim., ***Nature*** 544, 340 (2017)

Rules of remote atomic interaction through 2D materials

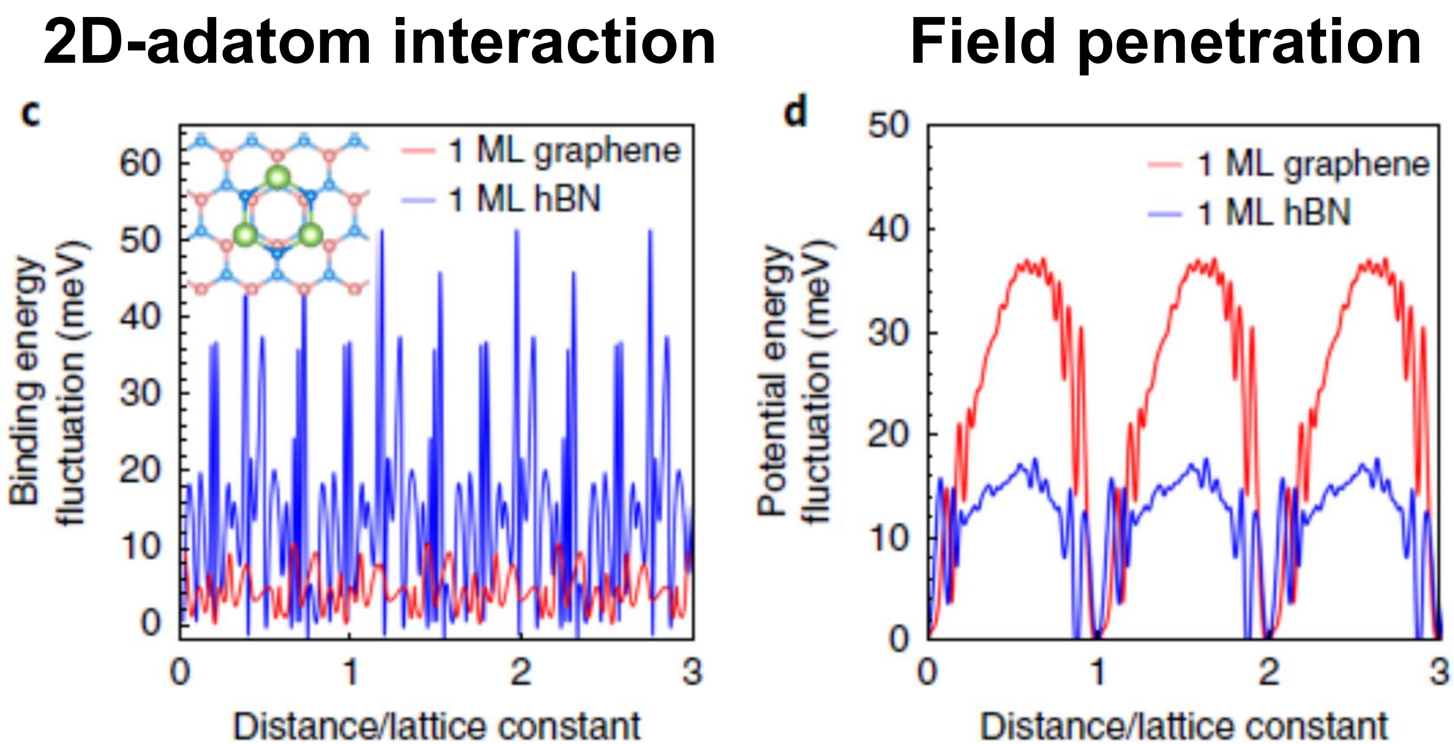


W. Kong,…and J. Kim*, ***Nature Materials***, Vol. 17, 335 (2018)

Polar vs non-polar (3D materials)

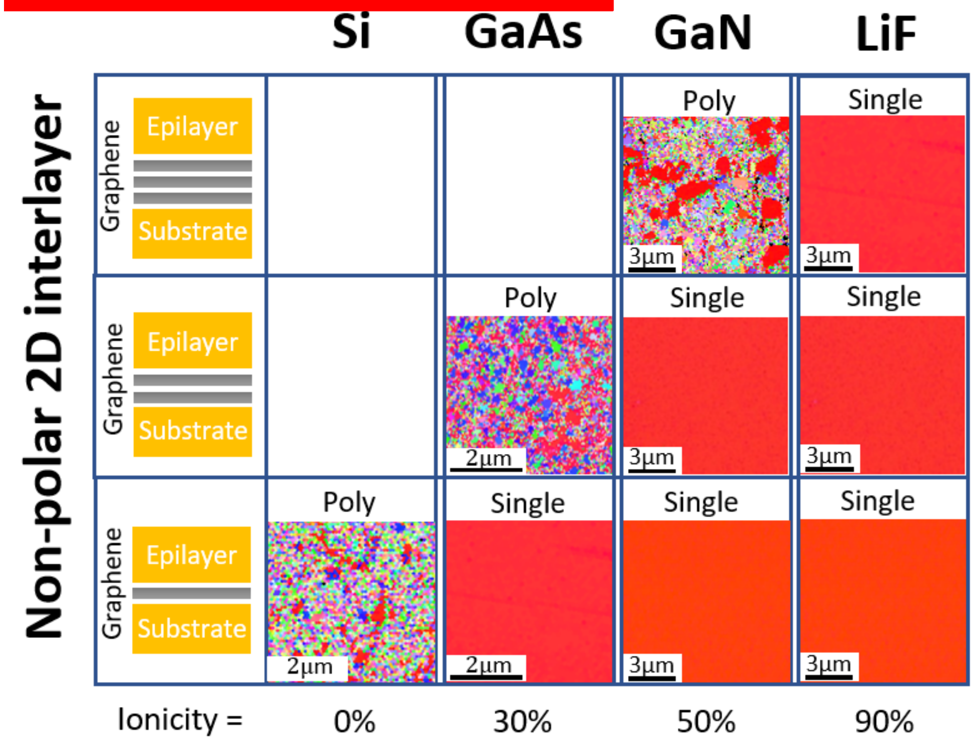


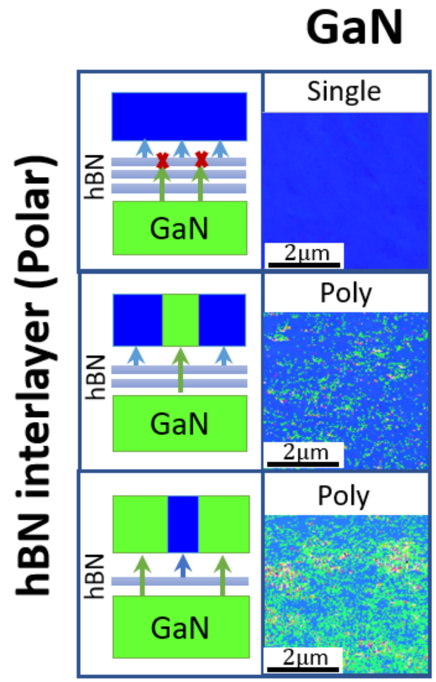
Polar vs non-polar (2D materials)

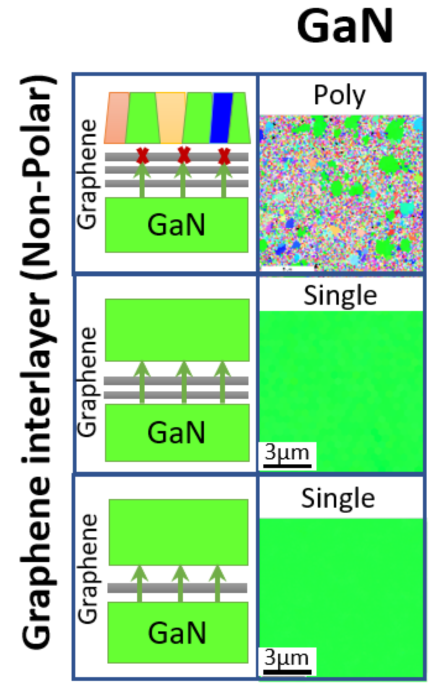


Polarity governs remote atomic interaction through 2D materials

DFT thermodynamic calculation

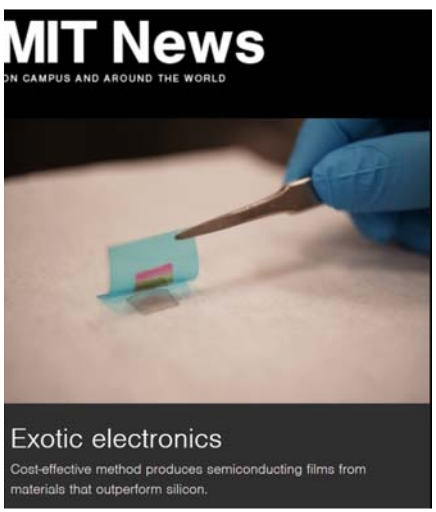






Polarity of 3D materials determines field penetration

Polarity of 2D materials determines field screening

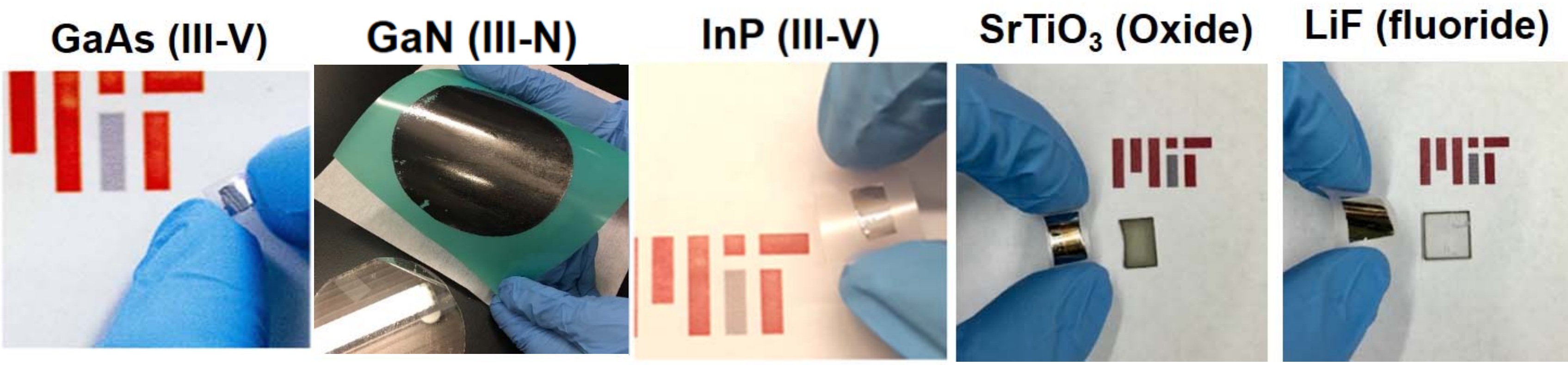


Y. Kim,… and J. Kim*., ***Nature*** 544, 340 (2017)

W. Kong,…and J. Kim*, ***Nature Materials***, Vol. 17, 335 (2018)

S. Bae…, and J. Kim*, ***Nature Materials***, To be published (2019)

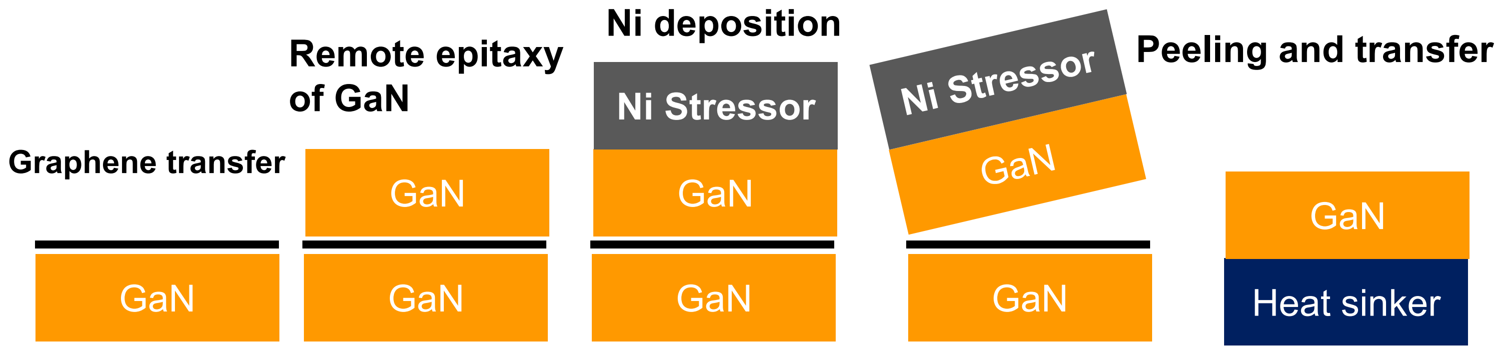
Technical impact of 2D material-based layer transfer

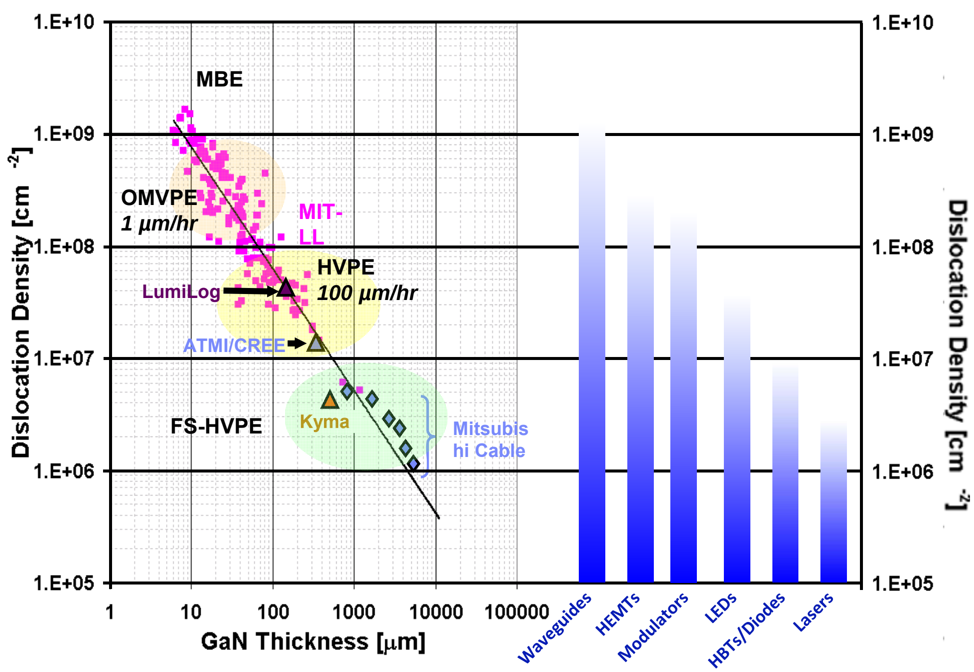


- **First demonstration of ultrafast wafer-scale layer release applicable for any materials**
  - UNLOCK historical technical barriers for electronics/optoelectronics/Energy storage
    - **For functional complex oxides**: Maximized ferroelectricity-ferromagneticity/Non epitaxial stacking
    - **For battery**: Dendrite-free single-crystalline electrolyte battery
    - **For III-V**: High efficiency solar cells with low cost via reusing wafers
    - **For Heterointegration:** Stacking of remote epitaxial layers
      - IoT sensors / MicroLEDs
      - Skin electronics: High performance all single-crystalline inorganic sensor arrays

Dislocation-free GaN by recycling FS-GaN wafer

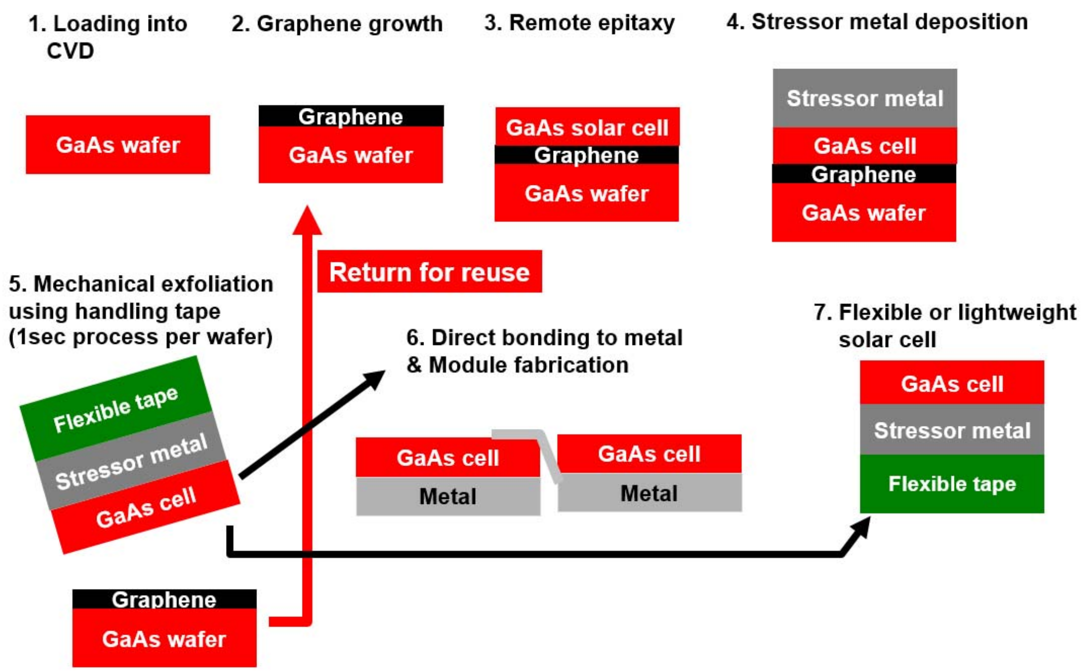
GaN on graphene/GaN



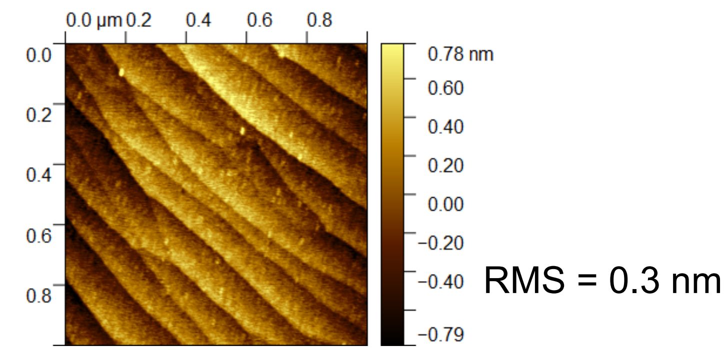


Spun-off company: Future Semiconductor Business

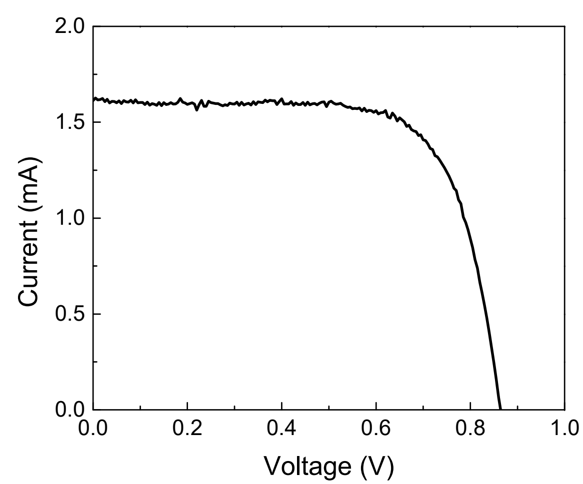
Wafer recycling for III-V solar cells



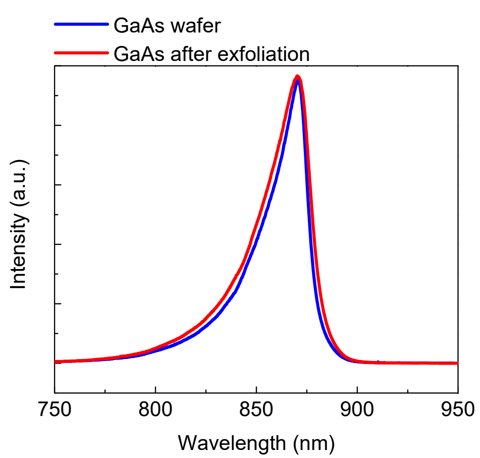
Smooth surface morphology of solar cells



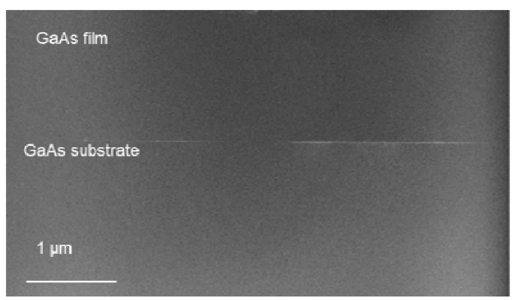
Photovoltaic response



Comparable PL spectra



Dislocation-free GaAs on graphene

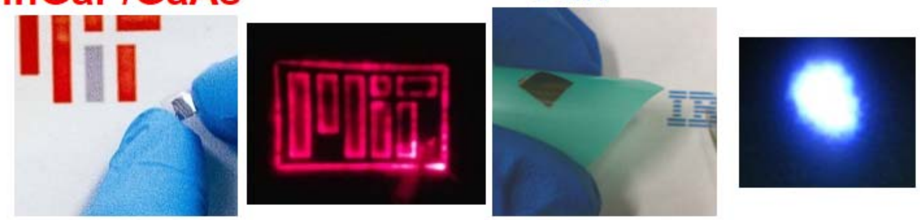


Masdar INSTITUTE
LG
U.S. DEPARTMENT OF ENERGY

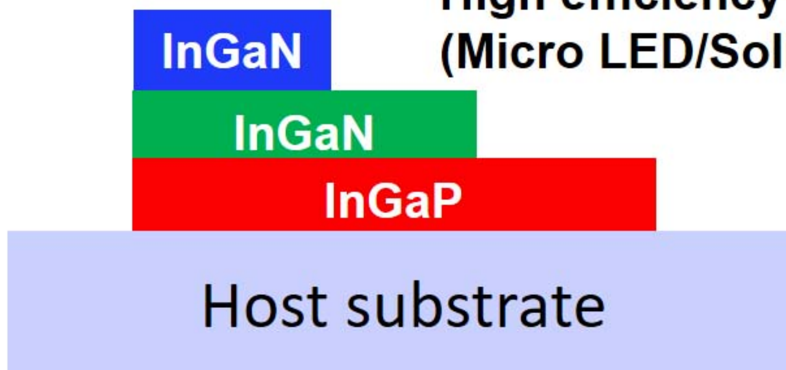
RGB full color microLED via remote epitaxy

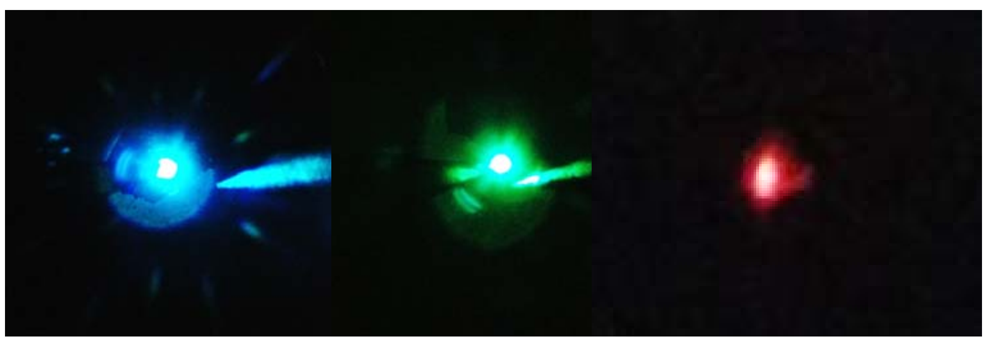
MicroLEDs

InGaP/GaAs GaN



High efficiency RGB
(Micro LED/Solid state lighting

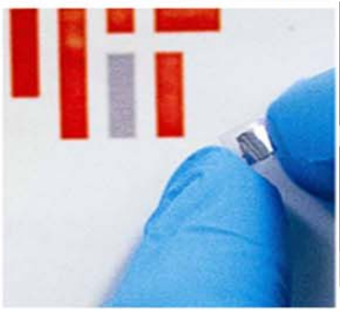




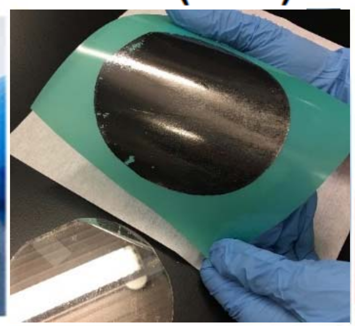
[remote epitaxially grown RGB pixels]

Ubiquitous electronics/IoT to be enabled by 2DLT

GaAs (III-V)



GaN (III-N)

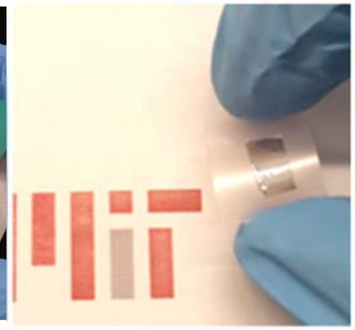


MicroLEDs

High efficiency PV
Transistor

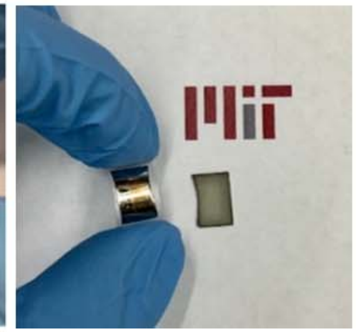
Wireless system
Power system

InP (III-V)



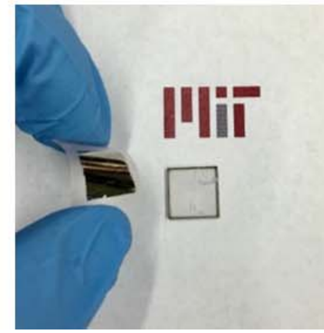
Photodetector

$SrTiO_3$ (Oxide)

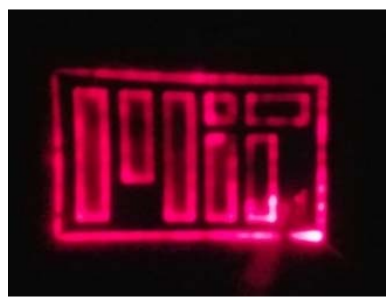


Piezoelectric sensor

LiF (fluoride)



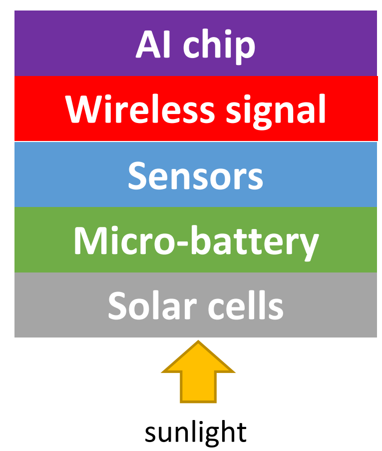
Optical waveguide
Battery



Monolayer graphene + Wafer

= Copy Machine (Film producer)

Self-powered/analyzed electronic wireless system



Y. Kim et al., and Jeehwan Kim., ***Nature*** 544, 340 (2017)

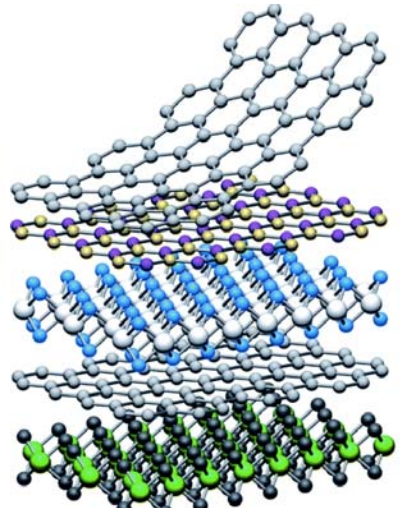
3D heterostructures enabled

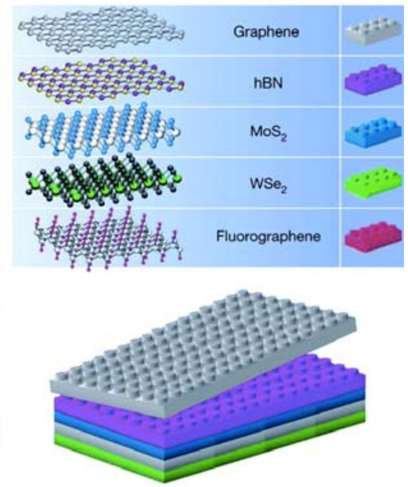
Many functions at one place!

Multi-functional devices by heterostructures

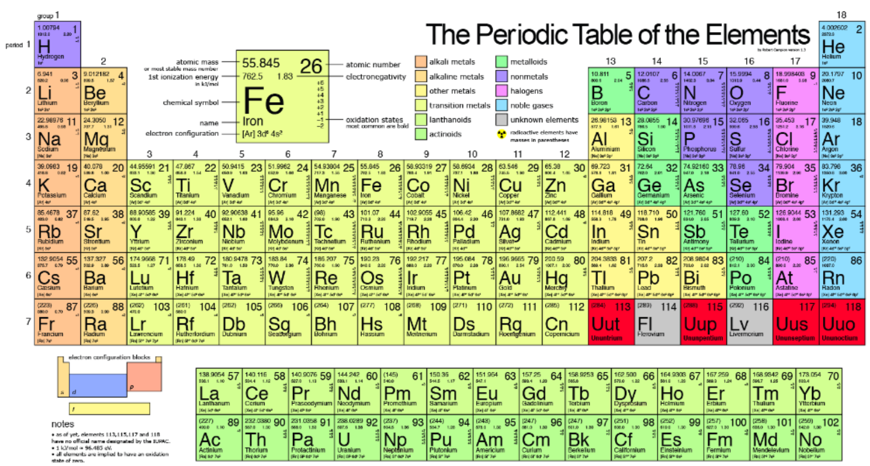
2D

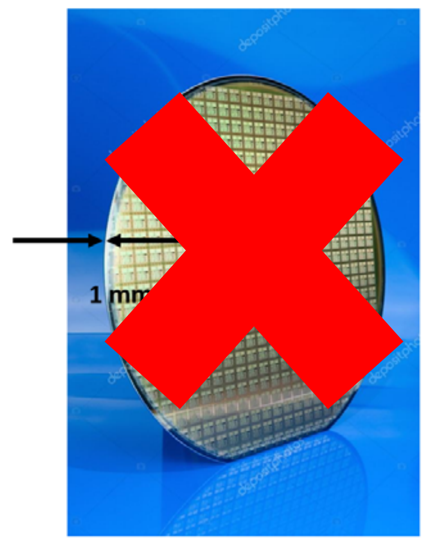
Graphene family	Graphene	hBN 'white graphene'	BCN	Fluorographene	Graphene oxide
2D chalcogenides	MoS_2, WS_2, $MoSe_2$, WSe_2	Semiconducting dichalcogenides: $MoTe_2$, WTe_2, ZrS_2, $ZrSe_2$ and so on		Metallic dichalcogenides: $NbSe_2$, NbS_2, TaS_2, TiS_2, $NiSe_2$ and so on Layered semiconductors: GaSe, GaTe, InSe, Bi_2Se_3 and so on	
2D oxides	Micas, BSCCO	MoO_3, WO_3	Perovskite-type: $LaNb_2O_7$, $(Ca,Sr)_2Nb_3O_{10}$, $Bi_4Ti_3O_{12}$, $Ca_2Ta_2TiO_{10}$ and so on	Hydroxides: $Ni(OH)_2$, $Eu(OH)_2$ and so on	
	Layered Cu oxides	TiO_2, MnO_2, V_2O_5, TaO_3, RuO_2 and so on		Others	





3D





Maximizing functionality by having ultrathin 3D & 2D

Extended material library: 2D or 3D → 2D & 3D

Extended structural configuration: 2D-2D or 3D-3D
→ 2D-3D, 3D-2D, 3D-2D-3D

2D on 3D

Transfer or
Van der Waals epitaxy

3D on 2D

Van der Waals epitaxy
or remote epitaxy

3D-2D-3D

Remote epitaxy

Massachusetts Institute of Technology

Wafer-scale 2D materials and their heterostructure

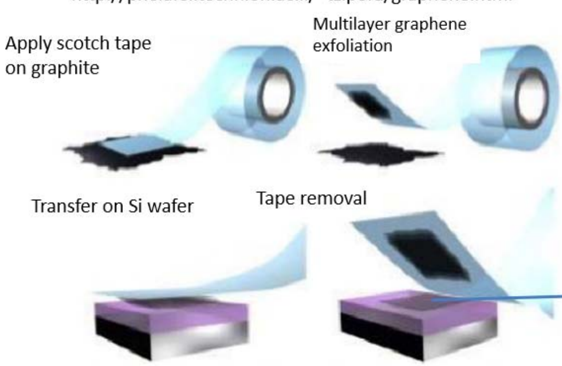
Jeehwan Kim
Research Group
http://jeehwanlab.mit.edu

#1 challenges in 2D material-based devices

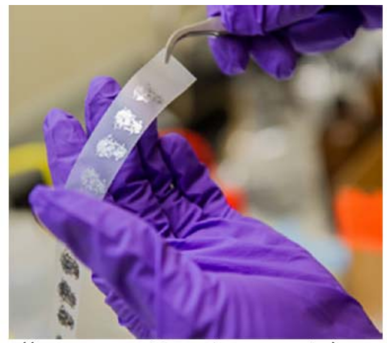
Small scale (tens of microns) / Monolayer isolation

Fabrication is still relying on scotch-tape flake method

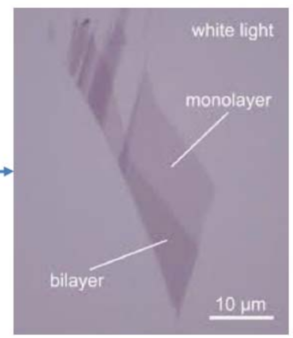
http://phelafel.technion.ac.il/~tzipora/graphene.html



Novoselov et al., Phys. Sci. 014006 (2012)



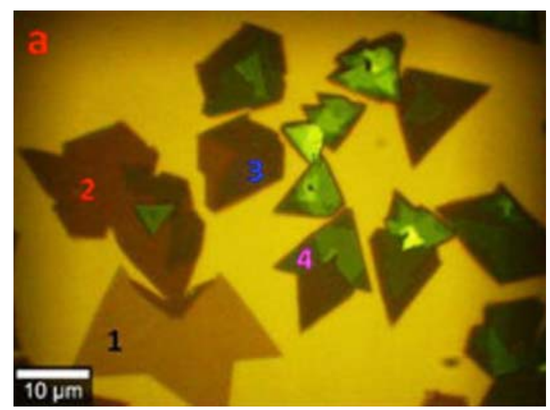
http://www.mccormick.northwestern.edu/magazine/



http://grapheneindustries.com

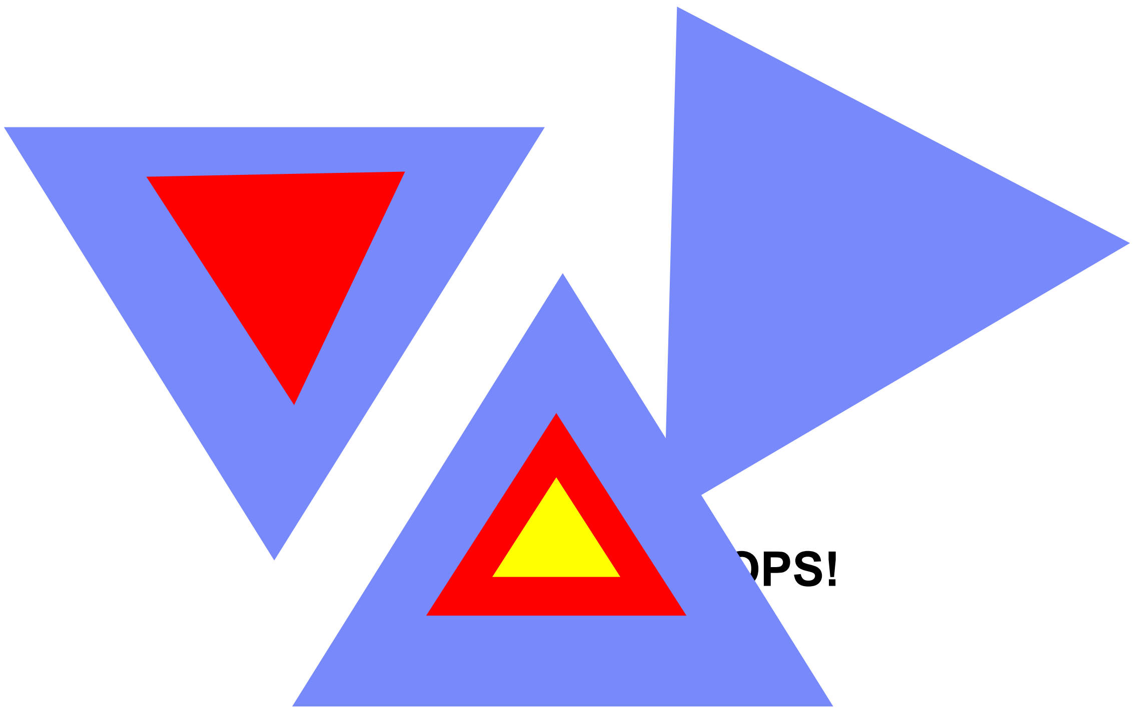
> 100 times repetition required for 1ML by 1 ML stacking

CVD growth cannot form wafer-scale monolayers



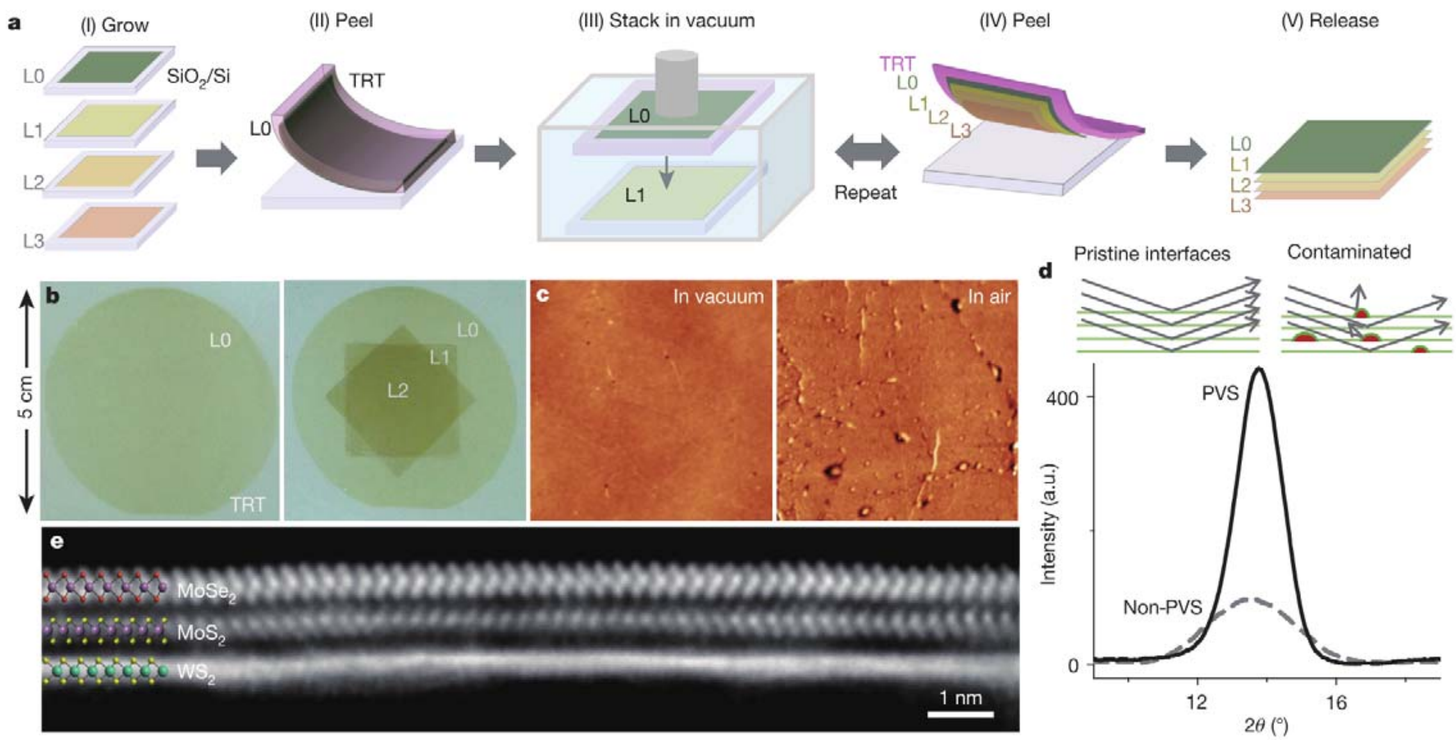
•*doi:10.1038/srep19476*

Challenge 2: Monolayer control (Growth)



Irregular thickness after growths

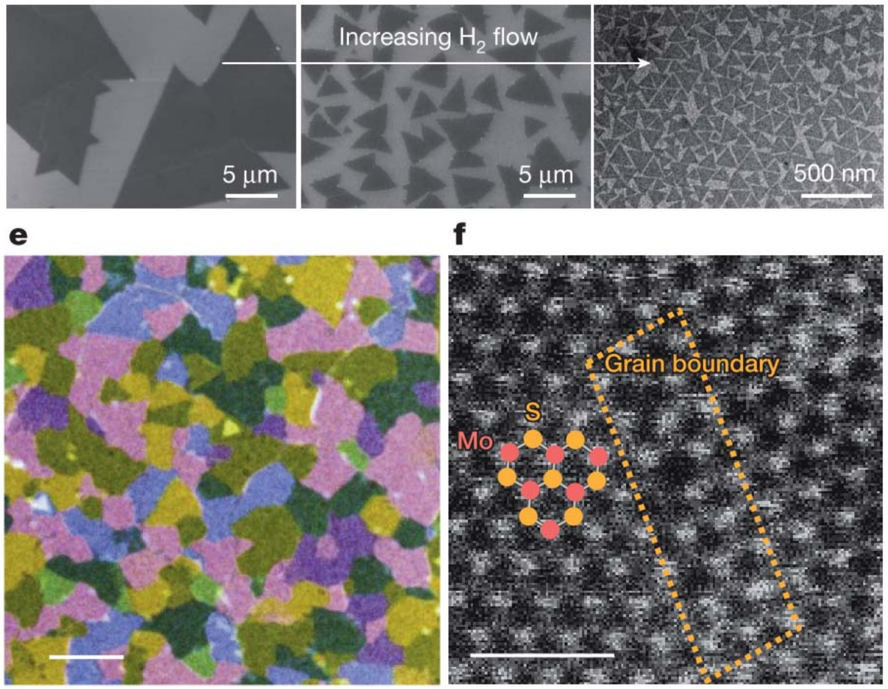
Challenge 3: Transferrability

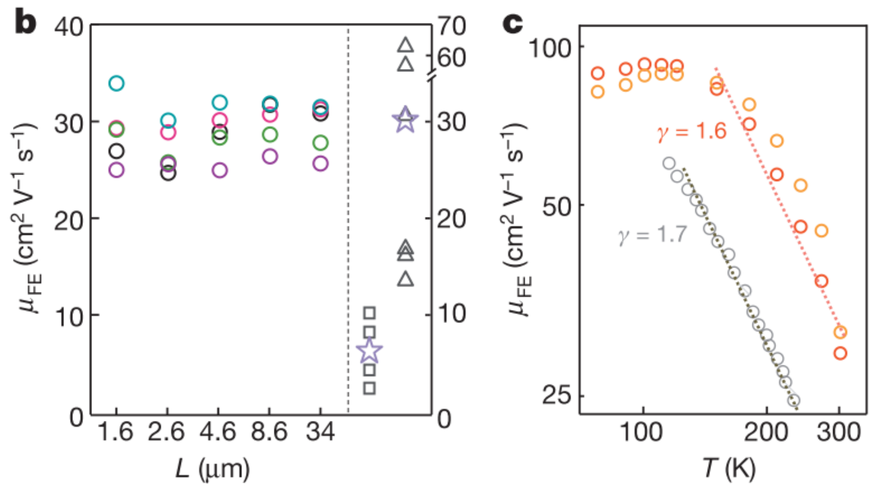


Nature 550 (2017)

Wafer-scale high yield peeling done if interaction with substrate is weak

Challenge 4: Low mobility due to small grain size





Nature 520 (2015)

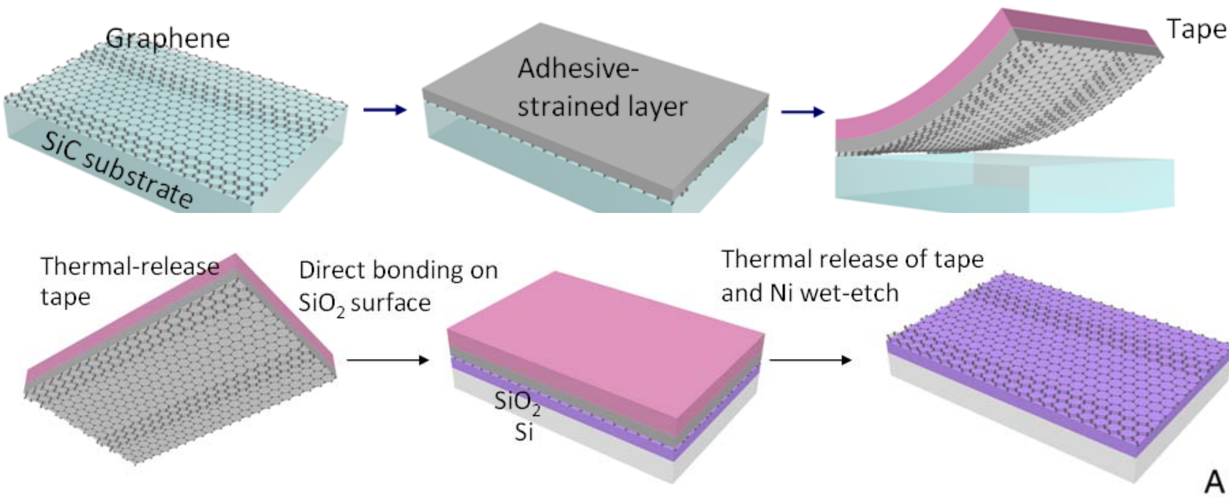
Grain size < 1 µm

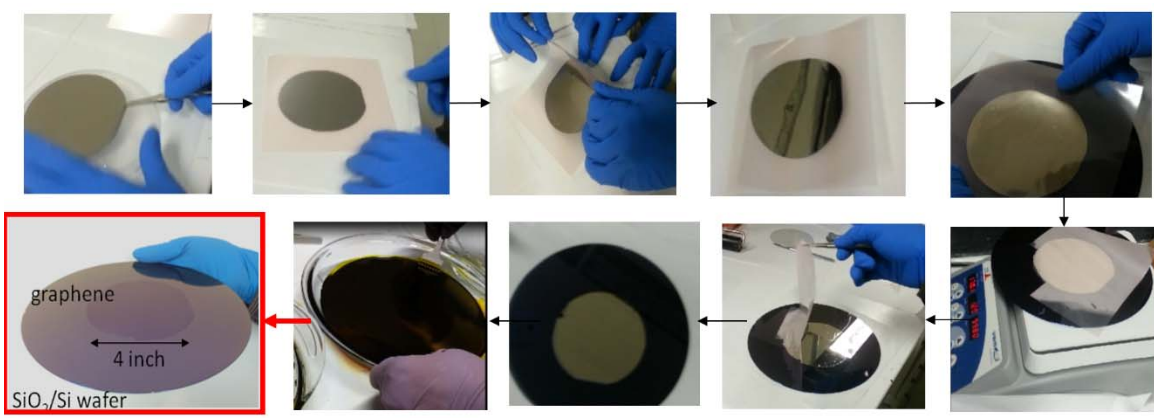
Mobility < 100 cm²/V-s

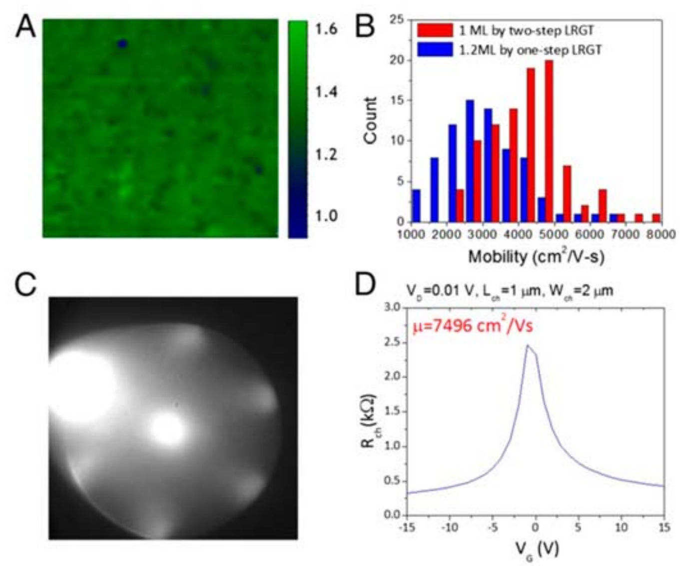
For transferability: Layer resolved graphene peeling and transfer

High yield 2D-3D interface separation technique

→ Using Ni as an atomic adhesive as well as a stressor







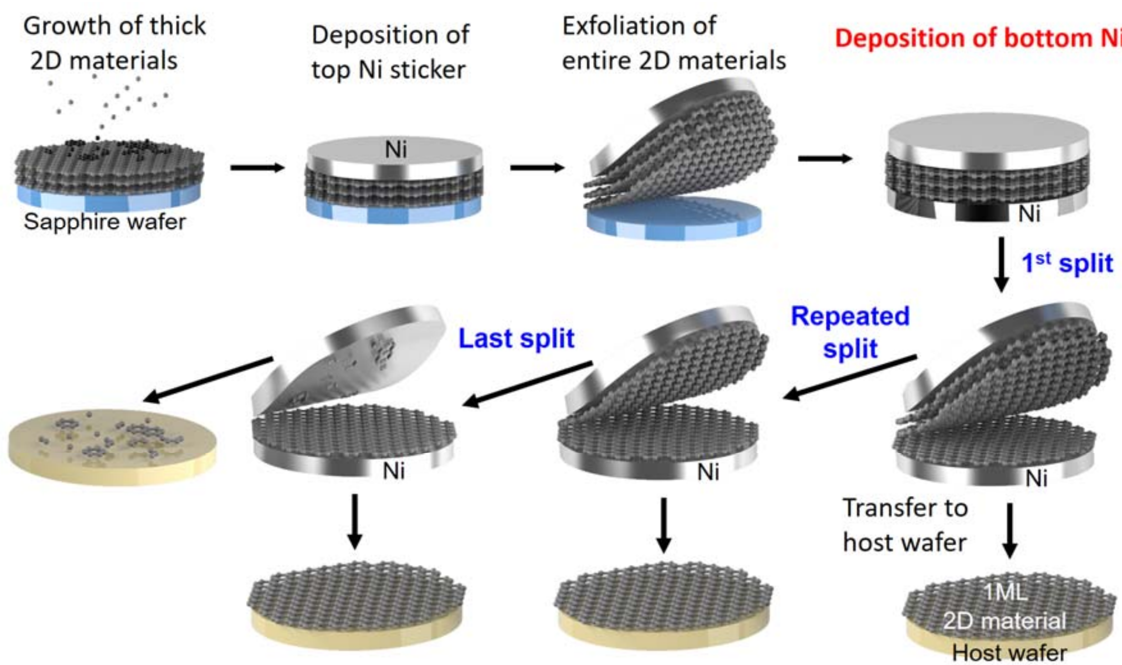
J. Kim et al. ***Science,*** 342, 833 (2013)

J. Kim et al. ***PNAS*** (2017)

For monolayer: Layer resolved splitting of 2D materials

Split multilayer 2D material wafers into many monolayers

→ *Using Ni as an atomic adhesive*

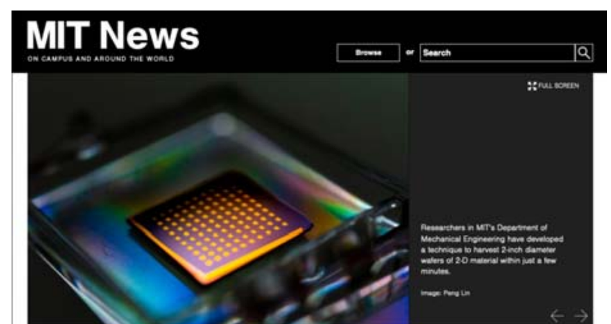


Science REPORTS

Cite as: J. Shim *et al.*, *Science* 10.1126/science.aat8126 (2018).

Controlled crack propagation for atomic precision handling of wafer-scale two-dimensional materials

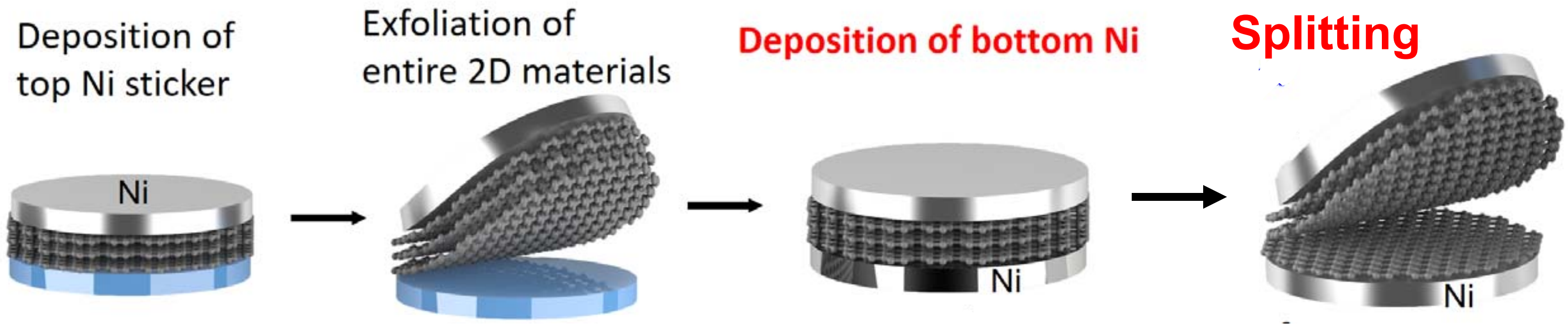
Jaewoo Shim[1,2*], Sang-Hoon Bae[1,2*], Wei Kong[1,2*], Doyoon Lee[1,2*], Kuan Qiao[1,2], Daniel Nezich[3], Yong Ju Park[4], Ruike Zhao[1,5], Suresh Sundaram[6], Xin Li[6], Hanwool Yeon[1,2], Chanyeol Choi[1,2], Hyun Kum[1,2], Ruoyu Yue[7], Guanyu Zhou[7], Yunbo Ou[8], Kyusang Lee[1,2,9], Jagadeesh Moodera[8], Xuanhe Zhao[1], Jong-Hyun Ahn[4], Christopher Hinkle[7,10], Abdallah Ougazzaden[6], Jeehwan Kim[1,2,11,12†]

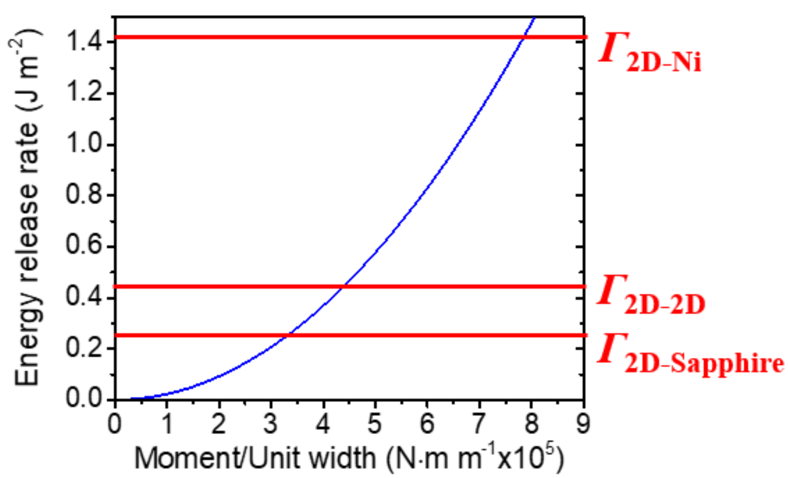


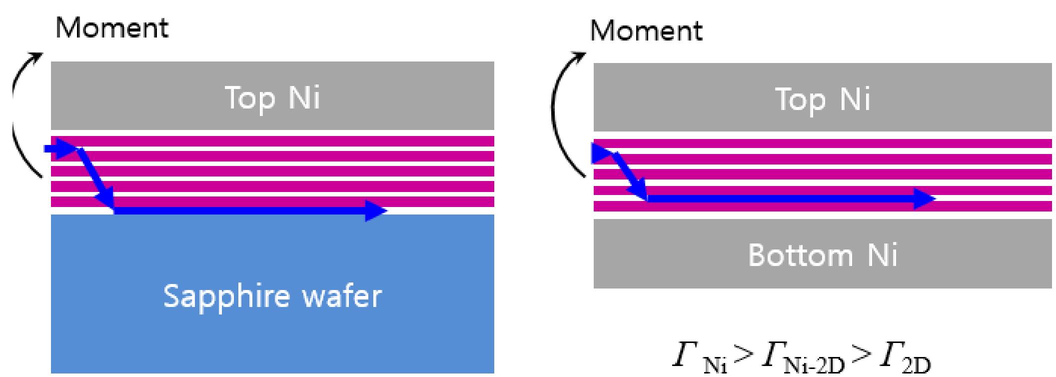
Researchers quickly harvest 2-D materials, bringing them closer to commercialization

Efficient method for making single-atom-thick, wafer-scale materials opens up opportunities in flexible electronics.

Crack control by playing with binding energy contrast



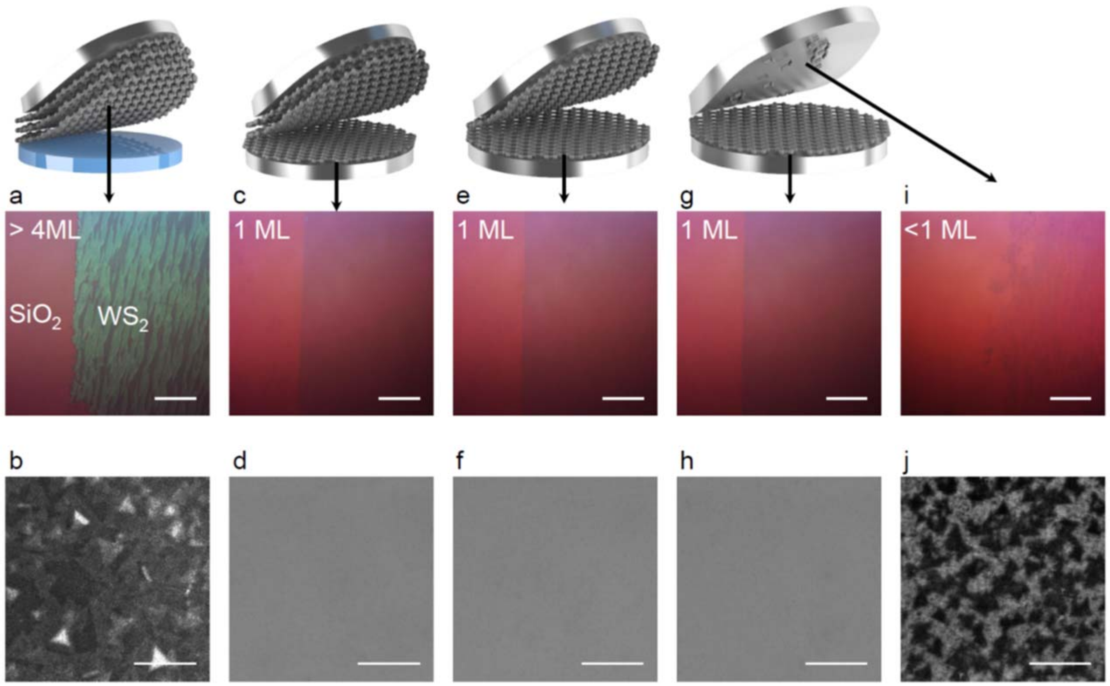


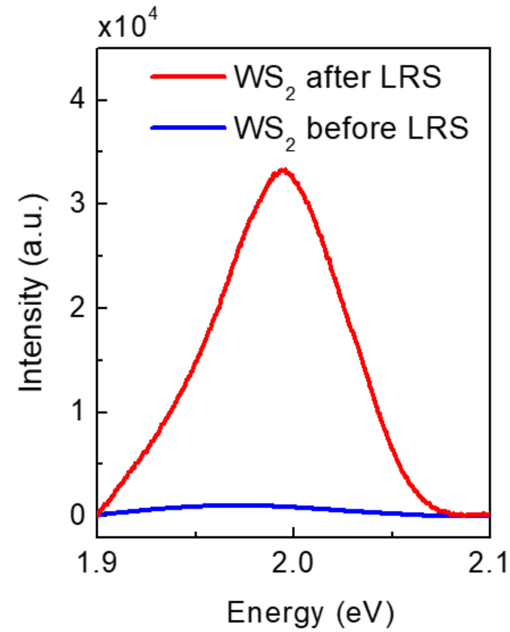


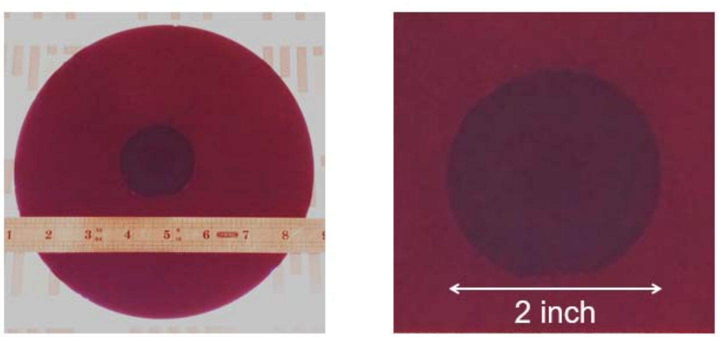
$\Gamma_{Sapphire} > \Gamma_{Ni} > \Gamma_{Ni-2D} > \Gamma_{2D} > \Gamma_{2D-Sapphire}$

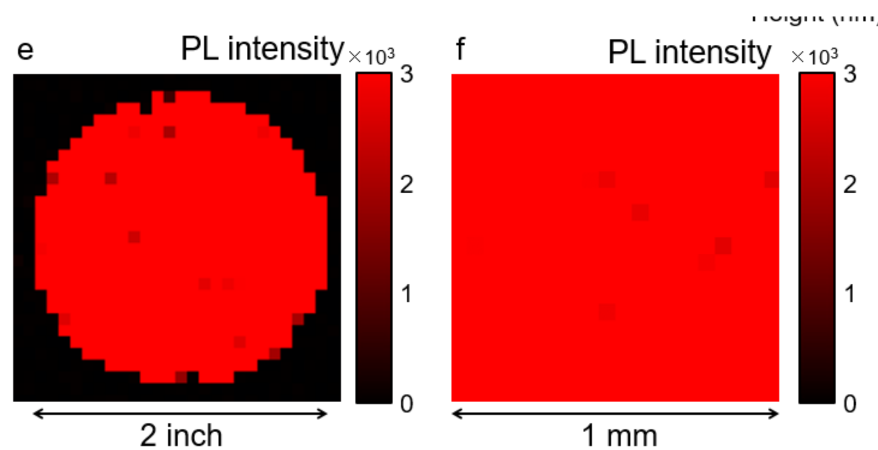
J. Shim,…and J. Kim*, ***Science*** (2018)

Three monolayers collected from 3 nm WS_2





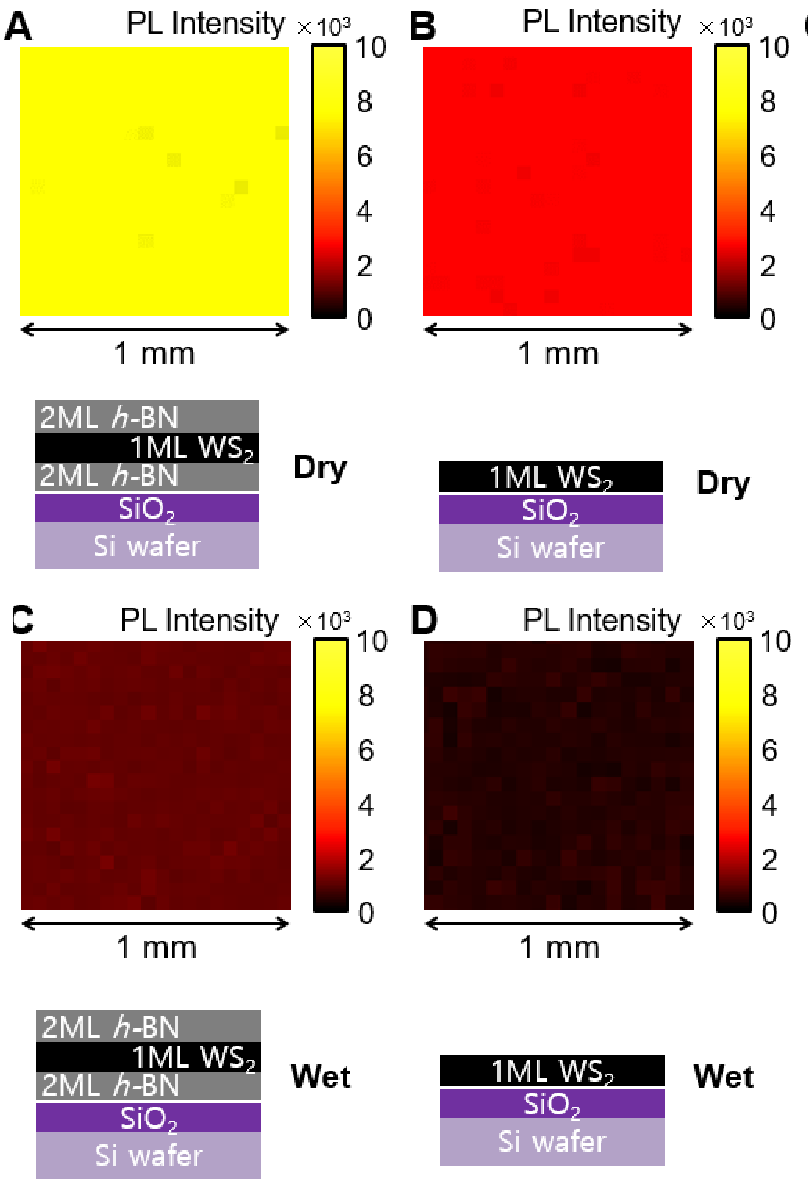


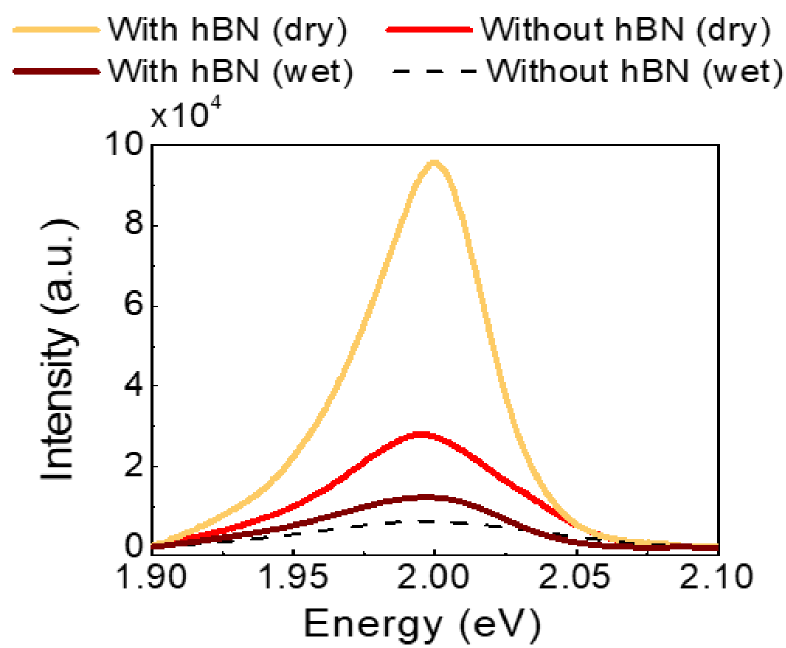


J. Shim,…and J. Kim*, ***Science*** (2018)

Monolayer isolation/stacking of hBN, TMDCs, graphene

Optoelectronic properties

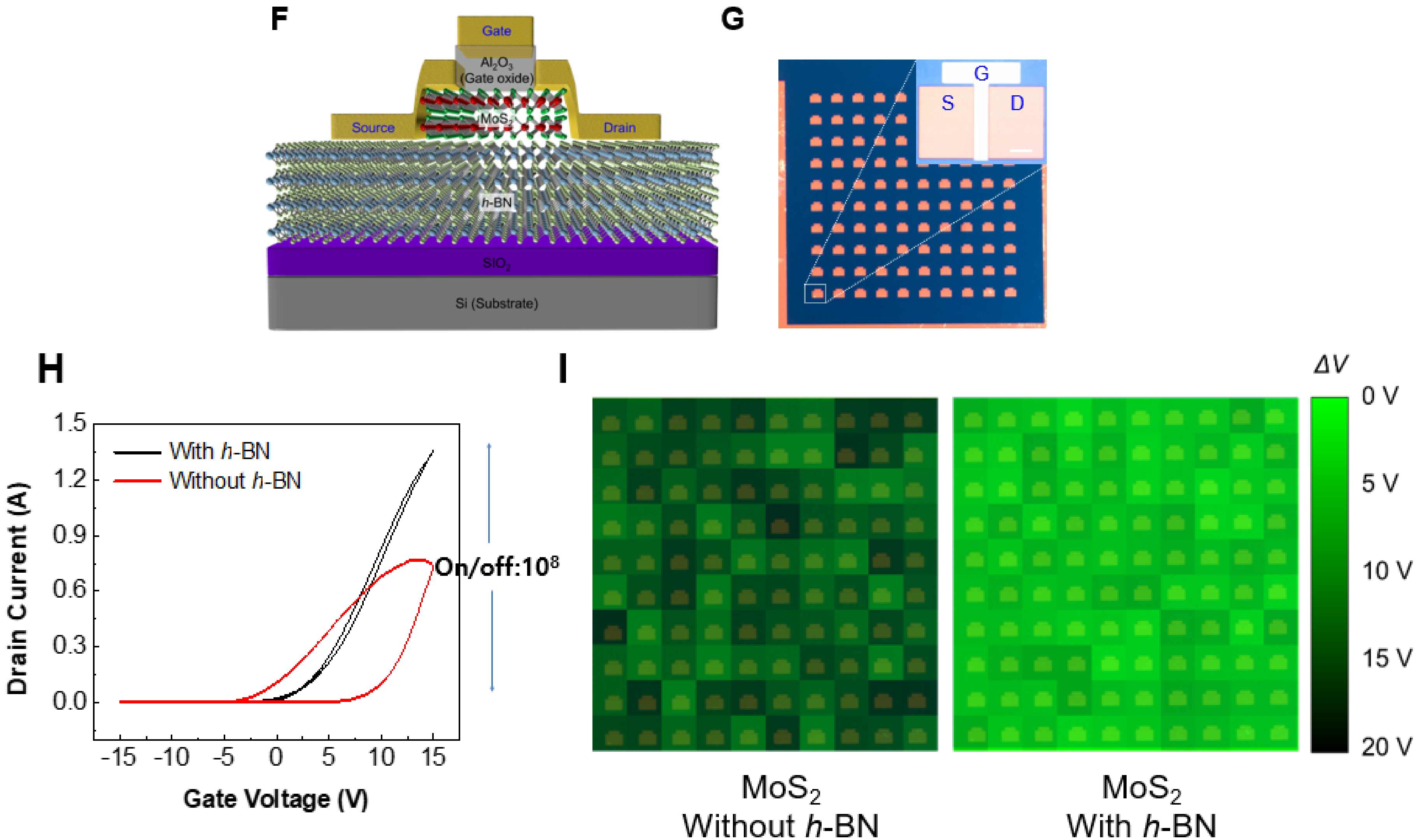




J. Shim,…and J. Kim*, ***Science*** (2018)

Application to hBN, TMDCs, graphene

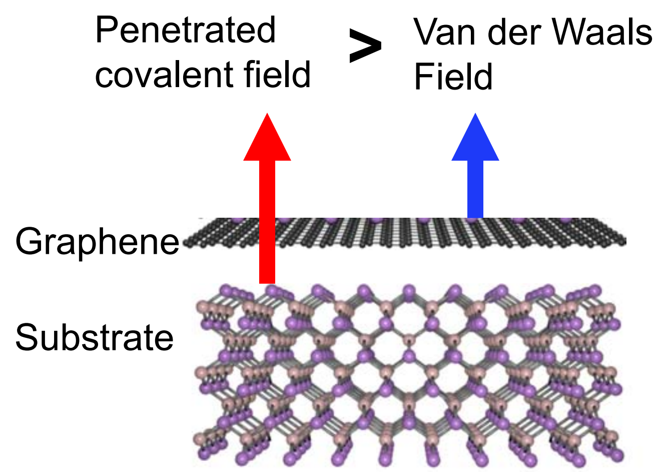
Uniform wafer-scale transistor arrays

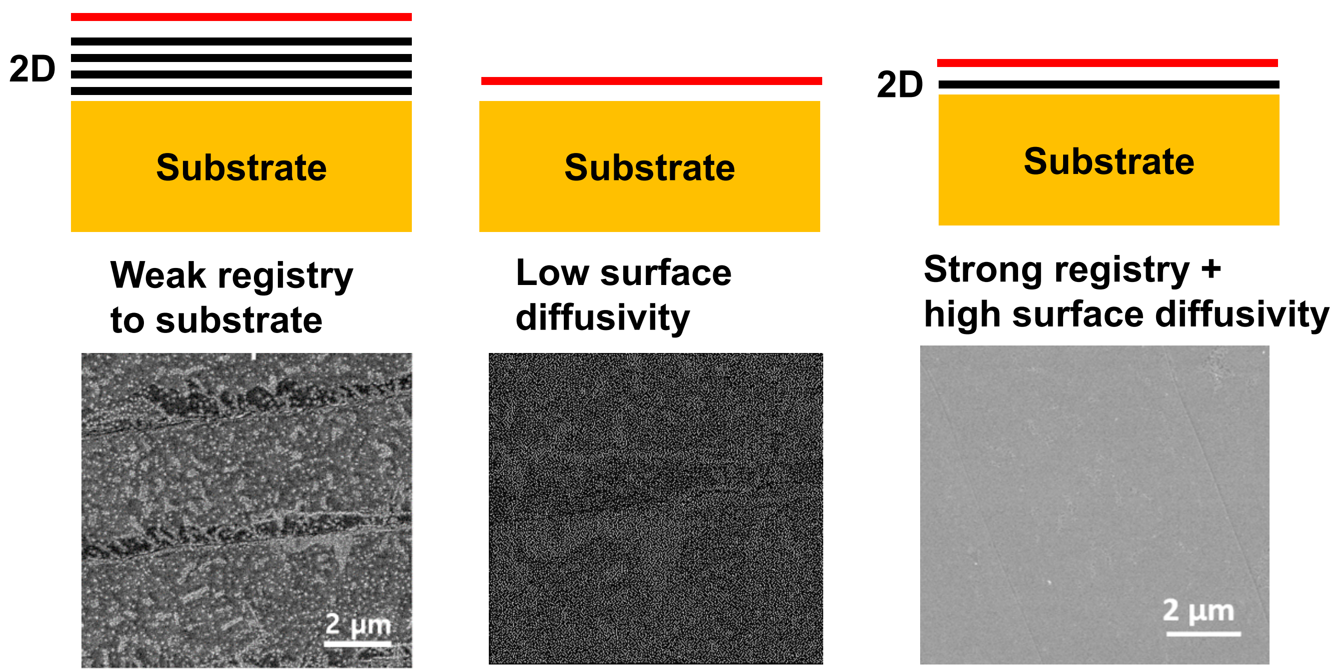


J. Shim,…and J. Kim*, ***Science*** (2018)

For mobility: Remote epitaxy of 2D materials

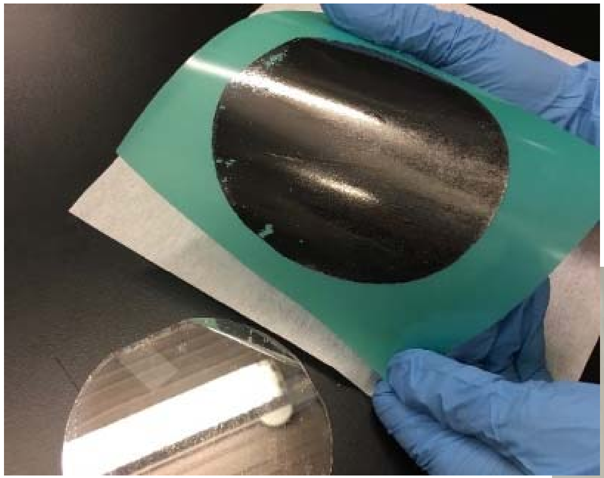
***Remote epitaxy** offers monolayer single-crystalline growth*





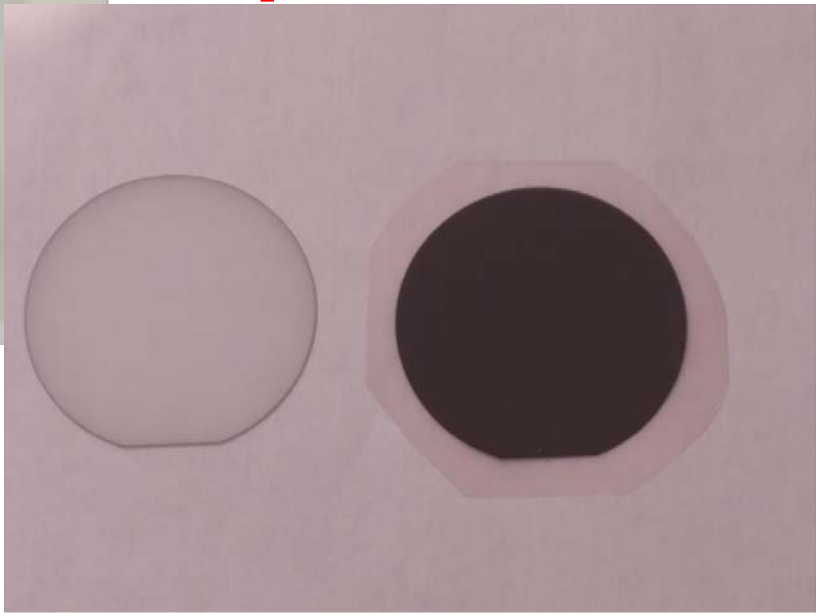
Wafer-scale tape process enabled for 3D and 2D materials

GaN peeled off from 4 inch wafer



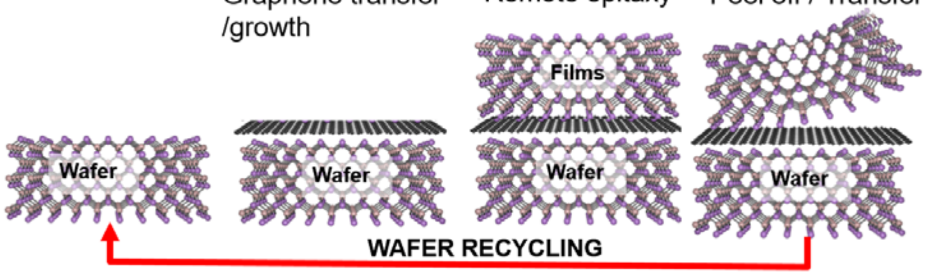
Graphene peeled off from 4 inch wafer

WS_2 peeled off from 4 inch wafer



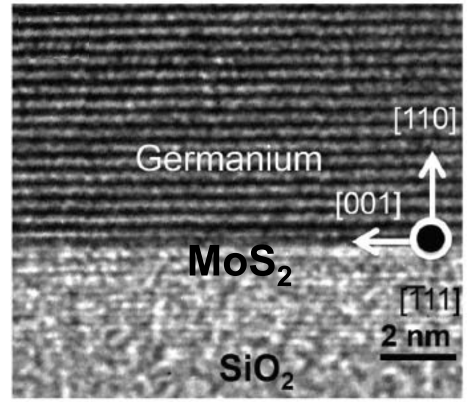
Implication of 3D-2D heterostructures

Van der Waals coupling



Cheap non-Si electronics

Electrical coupling

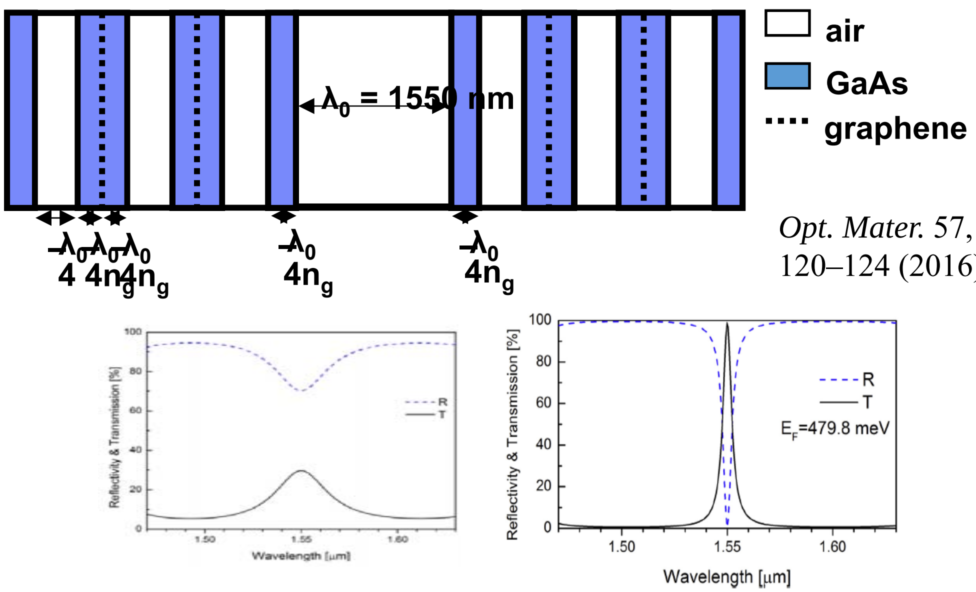


Charge transfer

Nanoscale 8, 18675–18681 (2016).

Carrier transfer between 3D-2D

Optical coupling

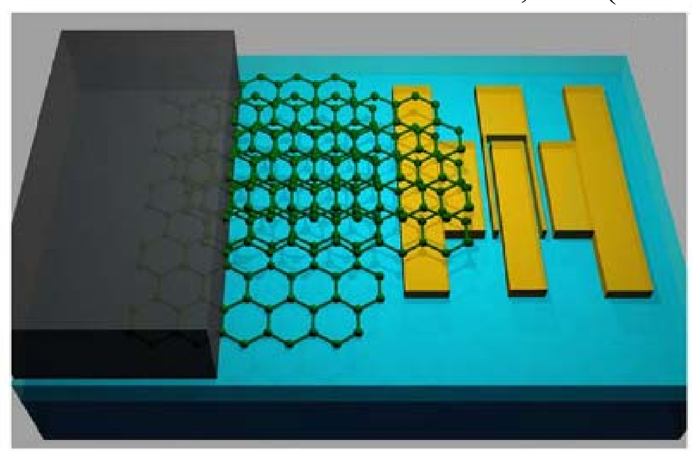


Opt. Mater. 57, 120–124 (2016)

Enhanced optics in 2D materials

Phonon coupling

Nat. Commun. 3, 827 (2012).



Heat transport from electronics

Massachusetts Institute of Technology

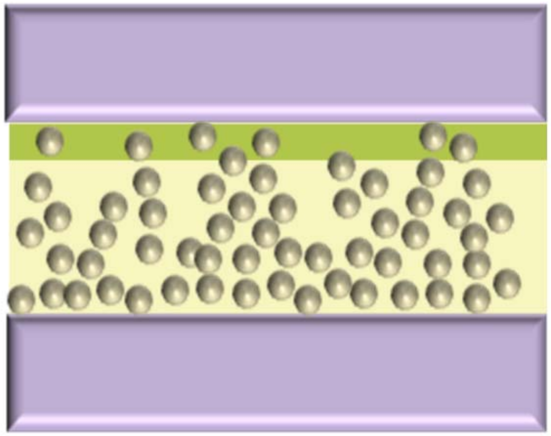
Team IV: Neuromorphic computing

Materials/Devices
ANN Arrays
Algorithm/Training

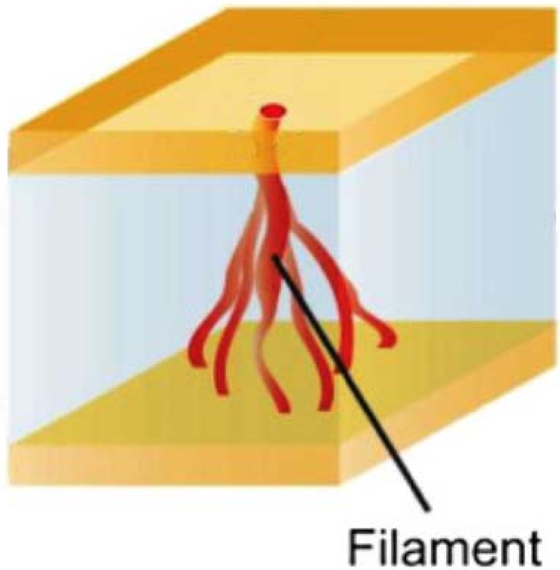
Materials/devices *ANN Arrays* *Algorithm/Training*

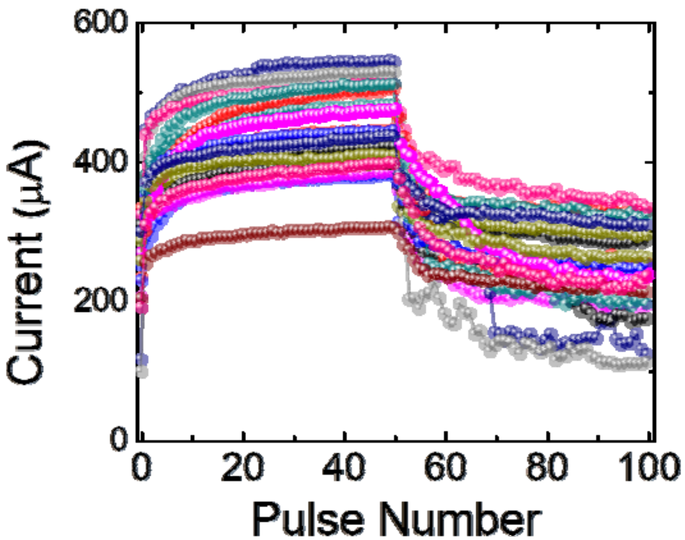
Unsolved problems in conventional memristors

Amorphous solid needed for defects to receive ions



Stochastic filament formation





S. Choi, et al.,Nano Letters (2017).

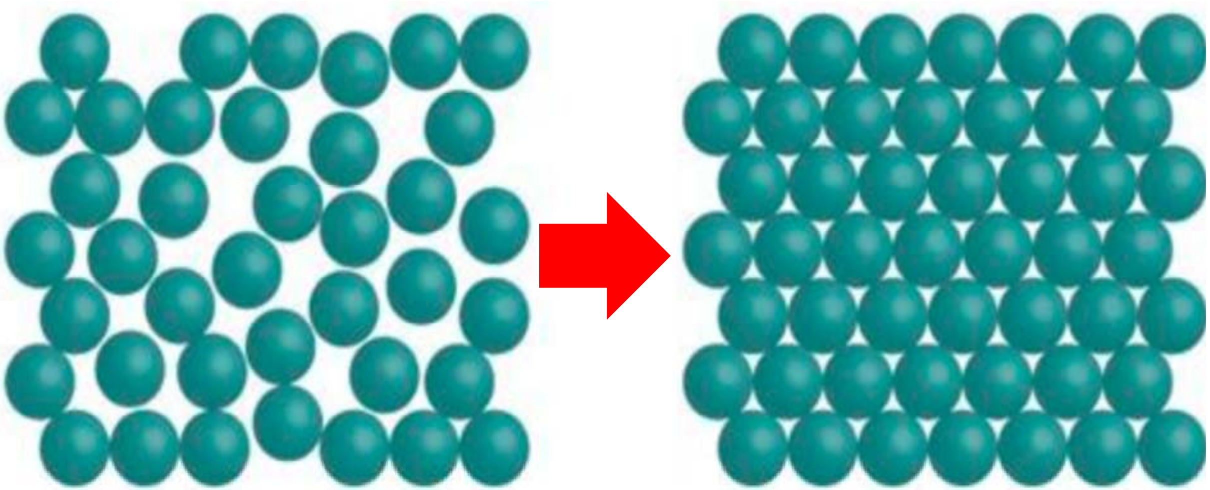
So many filament candidates
→ Source of variation

Pressurization of medium by ion injection
→ Source of non-linearity

Required memristor performance for neuromorphic computing

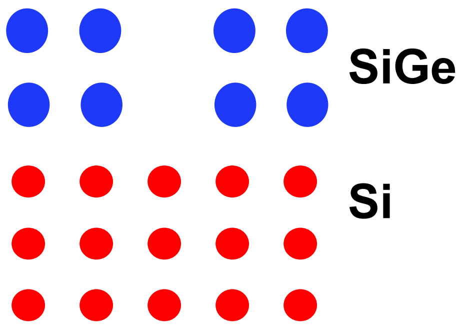
- Cycle-to-cycle uniformity
- Device-to-device uniformity
- High “analog” on-off ratio
- High endurance and long retention
- Symmetrical conductance update
- Current suppression in low voltage/reverse bias → Suppression of sneak paths

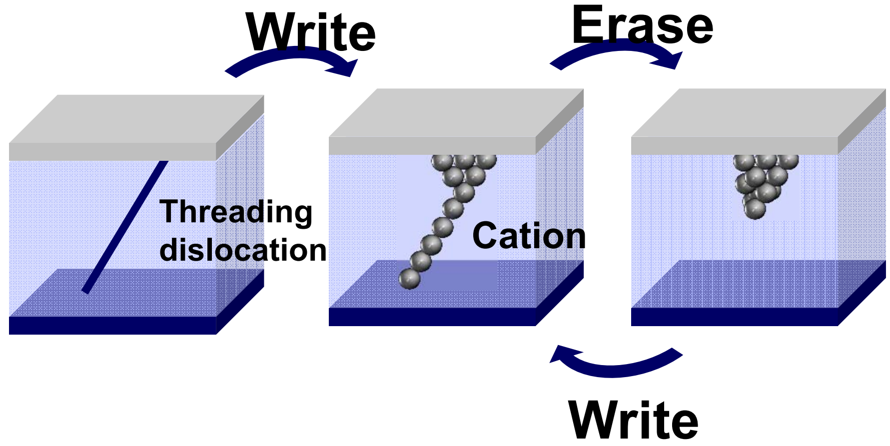
Why not starting from single-crystal?

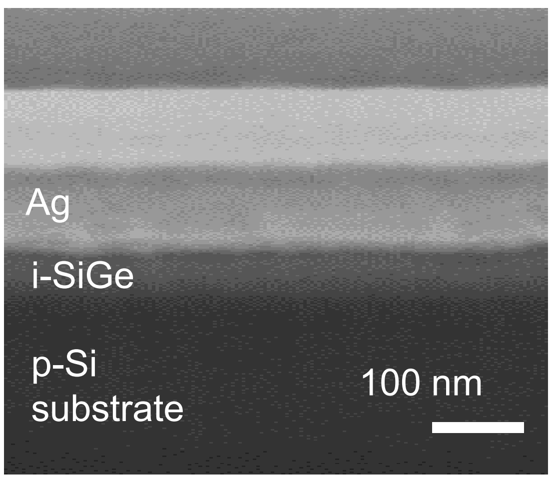


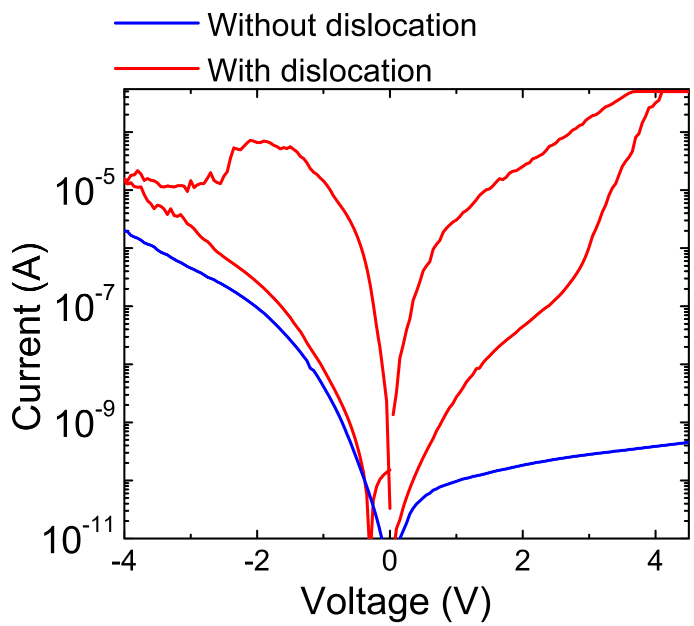
Perfect single crystal??
➔ no defect to receive ions

Epitaxial memory: Geometrical confinement





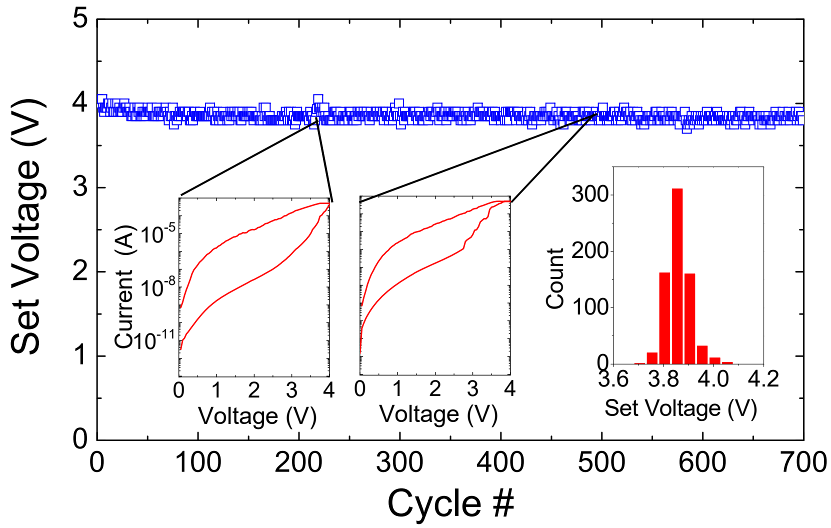




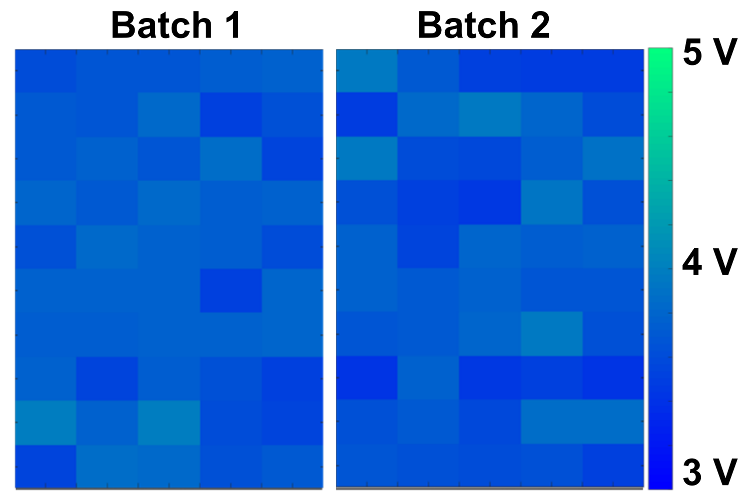
Dislocations in single-crystalline can be defined quantum paths of ion exchange

Consequence of confinement

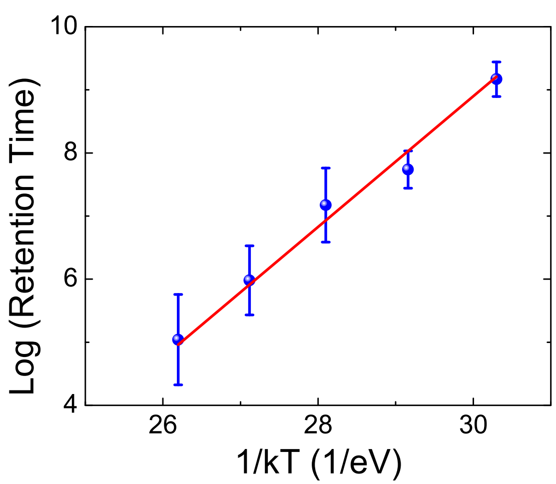
Temporal set V variation (cycle-to-cycle): 1%



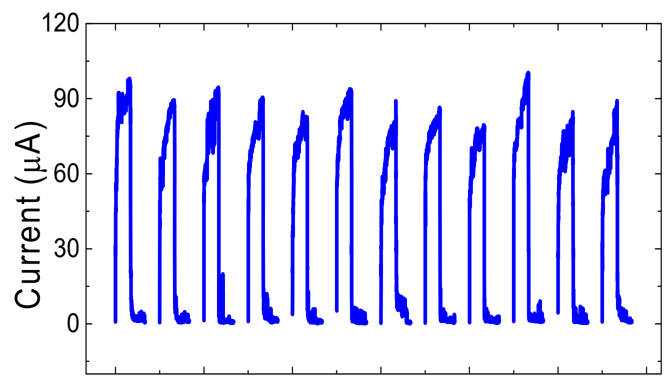
Spatial set V variation (device-to-device): 4%



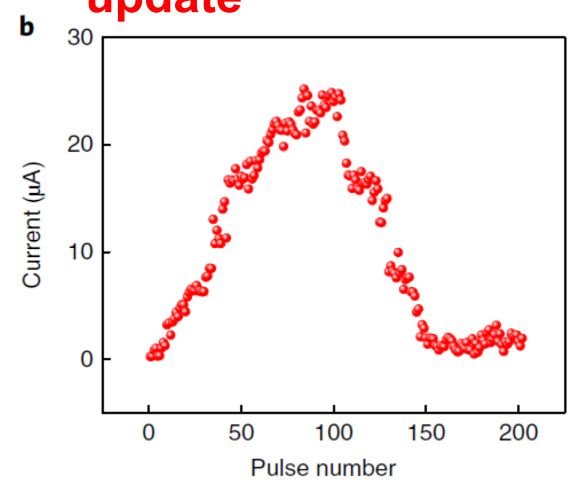
Long retention: 1.8 years at RT



Excellent Endurance : >10^9 pulses



Linear conductance update



Summary of epiRAM single device performance

Requirement for transistor-free operation of neuromorphic computing

- ❑ **High endurance and long retention** ✓
- ❑ **High on-off ratio** ✓
- ❑ **Cycle-to-cycle uniformity** ✓
- ❑ **Device-to-device uniformity** ✓
- ❑ **Suppression of sneak paths** ✓
- ❑ **Linear conductance update** ✓

Type	DC on/off (10^n)	Retention	Endurance (10^n cycles)	Set V Spatial Uniformity (σ/μ)	Set V Temporal Uniformity (σ/μ)	Reference
HfO_x	10	~ ms@ 27 °C	6	-	-	Wang 2016 PI: Joshua Yang, UMass
PEI/PEDOT:PSS	1	25 hr@ 85 °C	-	-	-	Burgt 2017 PI: Alberto Salleo, Stanford
Al_2O_3/TiO_x	4	14 hr@ 77 °C	3	0.11	-	Prezioso 2015 PI: Dimitri Strukov, UCSB
$Ag:CH_3NH_3PbI_3$	6	3 hr@ 27 °C	2	-	-	Choi 2016 PI: Ho Won Jang, SNU, Korea
Ta_2O_{5-x}/TaO_{2-x}	1	2.8 hr@ 250 °C	12	-	-	Lee 2011 Samsung, Korea
Ag:a-Si	1	-	-	0.03	-	Kim 2012 PI: Wei Lu, UMich
ZnO	1	0.3 hr@ 27°C	2	-	0.06	Chang 2010 PI: Tai-Bor Wu, NTHU, Taiwan
TiO_x	1	2.8 hr@ 27 °C	2	0.10	-	Kim 2011 PI: Keon Jae Lee, KAIST, Korea
SiO_2	3	110 hr@ 85°C	-	0.10	-	Choi 2011 PI: I-Wei Chen, UPenn
WO_x	Analog	Decaying with ~s time constant(τ)	-	-	-	Chang 2011 PI: Wei Lu, UMich
SiGe epiRAM	**4**	**48 hr@ 85 °C**	**9**	**0.04**	**0.01**	**MIT**

nature materials | ARTICLES

https://doi.org/10.1038/s41563-017-0001-5

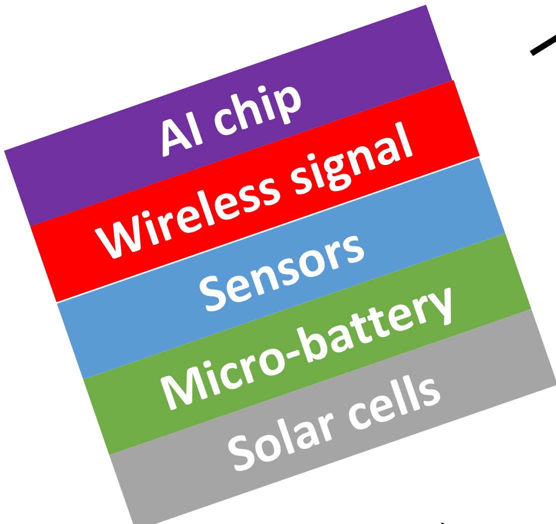
SiGe epitaxial memory for neuromorphic computing with reproducible high performance based on engineered dislocations

Shinhyun Choi[1,2], Scott H. Tan[1,2], Zefan Li[1,2], Yunjo Kim[1,2], Chanyeol Choi[1,2], Pai-Yu Chen[3], Hanwool Yeon[1,2], Shimeng Yu[3] and Jeehwan Kim[1,2,4]*

S. Choi, S. Tan, Y. Kim, C. Choi, P. Chen, S. Yu, and J. Kim*, "SiGe epitaxial memory for neuromorphic computing with reproducible high performance based on engineered dislocations", ***Nature Materials***, (2018)

Jeehwan Kim group at MIT: Electronics on demands

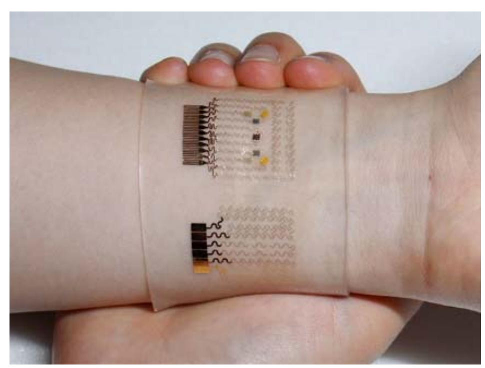
- Any electronic functions
- Any shapes
- Any places



Oil fields for smart oil extraction

Buildings for smart city

Skins for human health



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Remote Epitaxy for Energy

HYUNDAI
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AIR FORCE RESEARCH LABORATORY

Remote Epitaxy for Power

ANALOG DEVICES
ROHM SEMICONDUCTOR
DARPA
LINCOLN LABORATORY
MASSACHUSETTS INSTITUTE OF TECHNOLOGY

2D-based layer transfer

AMOREPACIFIC
NSF
Lam RESEARCH

Neuromorphic computing

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