Oct 31, 2014 at NTT Basic research Labs, Atsugi

Correlation between morphology and transport properties of quasi-free-standing monolayer graphene (QFMLG)

Yuya Murata[1], Torge Mashoff[2], Makoto Takamura[3], Shinichi Tanabe[3], Hiroki Hibino[3], Fabio Beltram[1,2], and Stefan Heun[1]

1 NEST, Istituto Nanoscienze-CNR and Scuola Normale Superiore, Pisa, Italy

2 Center for Nanotechnology Innovation @ NEST, Istituto Italiano di Tecnologia, Pisa, Italy

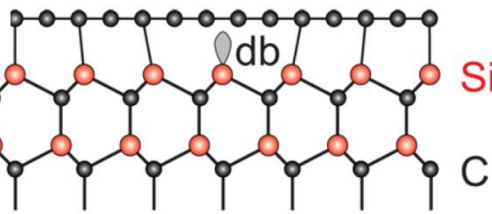
3 NTT Basic Research Laboratories, NTT Corporation, Japan

National Enterprise for nanoScience and nanoTechnology

Introduction

Graphene on silicon carbide (SiC) (0001)

buffer layer

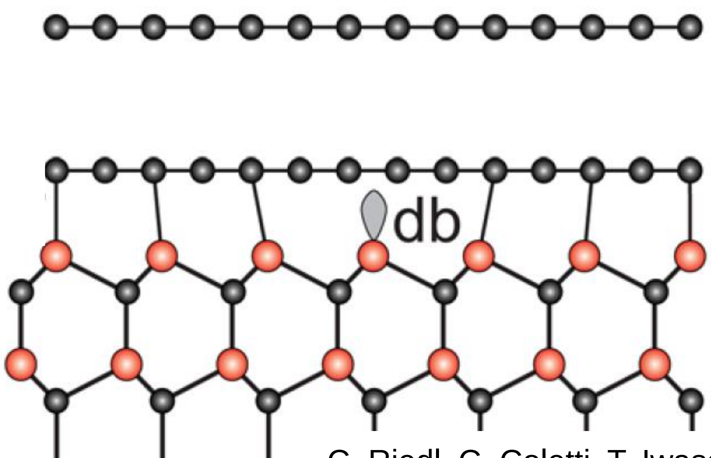


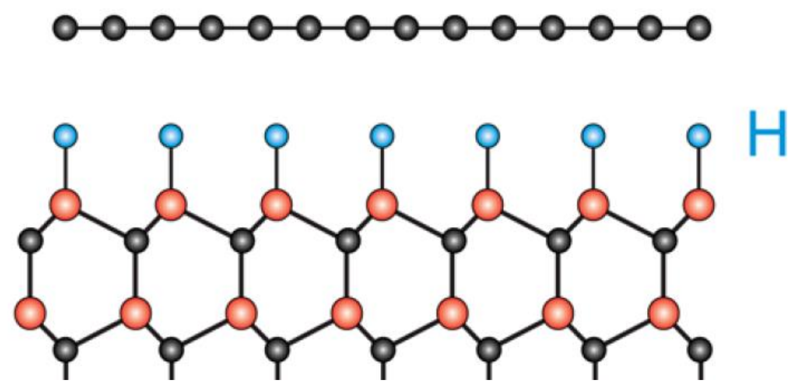
annealing

H intercalation

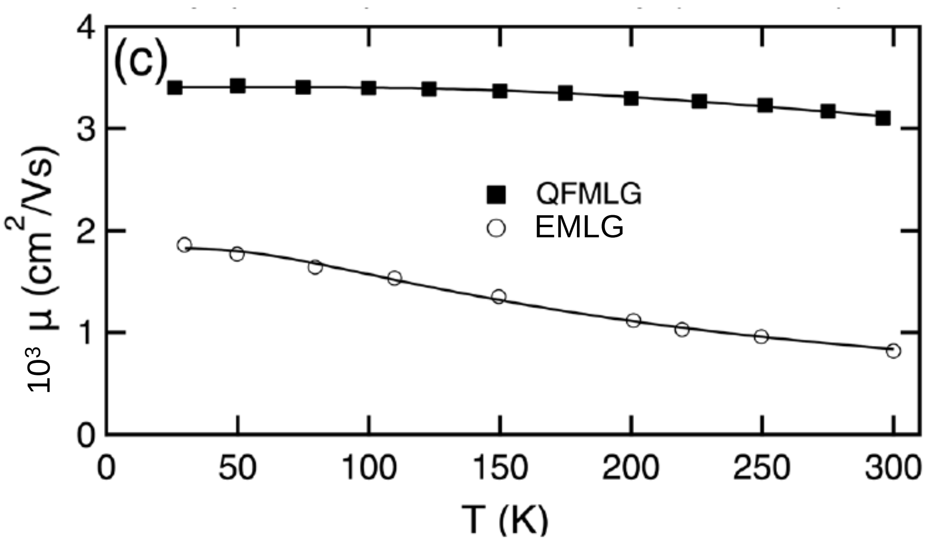
epitaxial monolayer graphene (EMLG)

quasi-free-standing monolayer graphene (QFMLG)





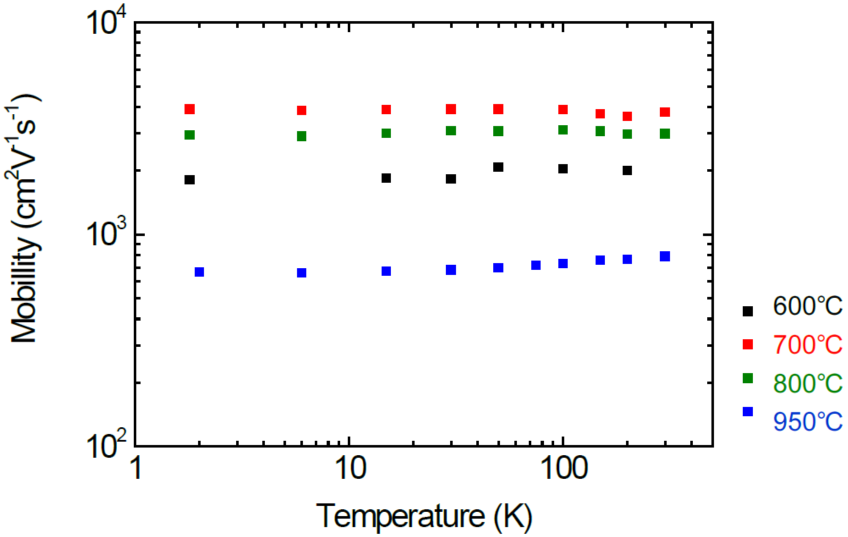
C. Riedl, C. Coletti, T. Iwasaki, A. A. Zakharov, and U. Starke, Phys. Rev. Lett. **103**, 246804 (2009)

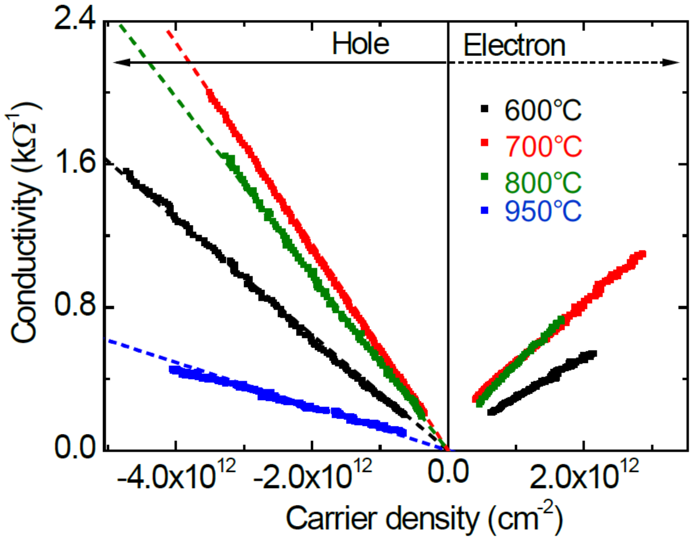


The carrier mobility of QFMLG shows less temperature dependence than EMLG, indicating less interaction between QFMLG and the SiC substrate.

However, the mobility of QFMLG (-3000 $cm^2V^{-1}s^{-1}$) is still lower than exfoliated graphene on SiO_2 or free standing graphene.

F. Speck, J.Jobst, F. Fromm, M. Ostler, D. Waldmann, M. Hundhausen, H. B. Weber, and Th. Seyller, Appl. Phys. Lett. **99**, 122106 (2011)





S. Tanabe, M. Takamura, Y. Harada, H. Kageshima, and H. Hibino, Jpn. J. Appl. Phys. **53**, 04EN01 (2014).

The QFMLG mobility depends on T_H,
the substrate temperature during H intercalation

highest mobility by T_H = 700ºC

conductivity – carrier density

- linear for T_H = 600-800ºC
  - charged impurity
- sublinear for T_H = 950ºC
  - additional scattering by defect

Purpose :
to observe the morphology of QFMLG formed at different T_H and investigate the relationship with transport property

Experiment

sample: 4H or 6H-SiC(0001)

cleaning anneal at 1500ºC for 5 min in H_2 of 33 mbar

buffer layer growth

anneal at 1650ºC for 5 min in Ar of 800 mbar

H intercalation

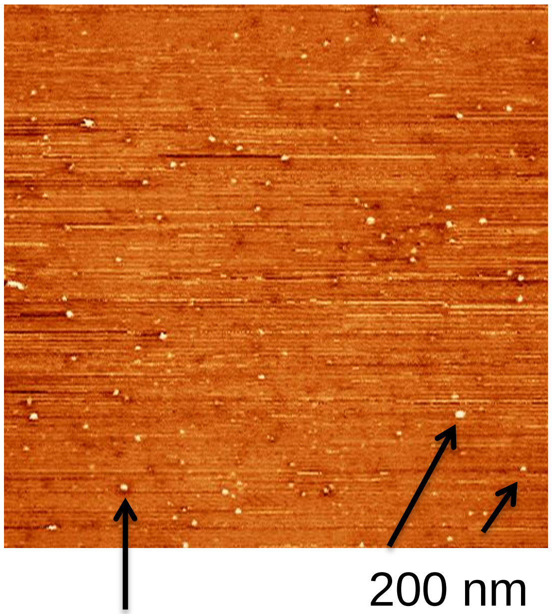
anneal at **<u>600 - 1200ºC</u>** for 1 hour in H_2 of 1 atm

characterization

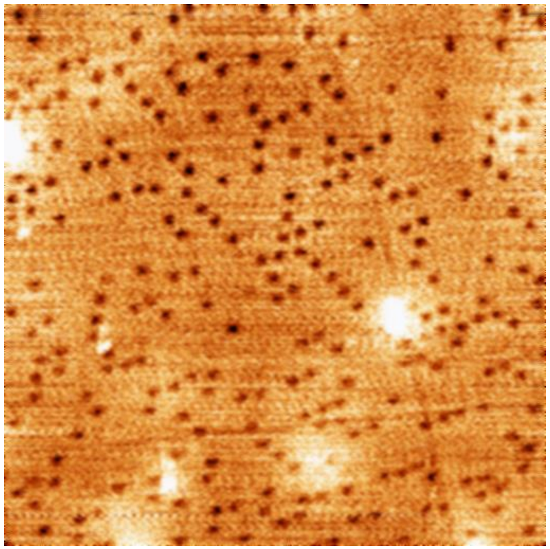
- STM in ultra-high vacuum (1×10^{-10} mbar)
- AFM in air
- TEM

National Enterprise for nanoScience and nanoTechnology

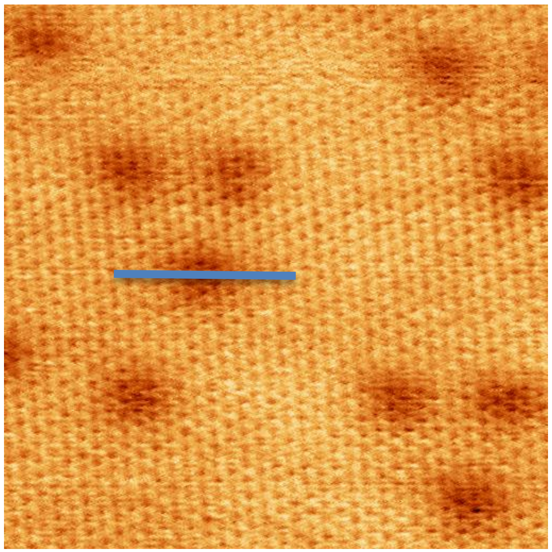
STM $T_H = 600^{o}C$



200 nm



50 nm



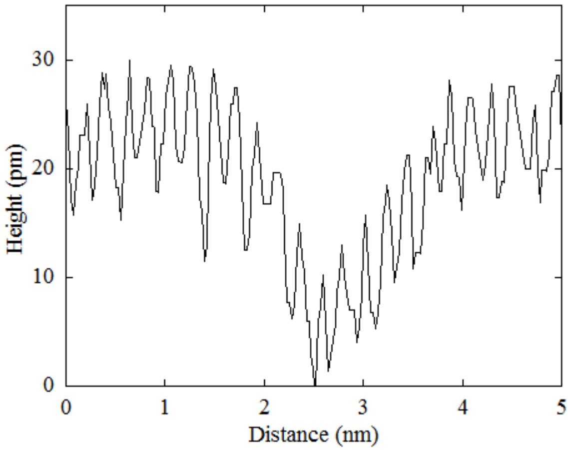
8 nm

- bright spots

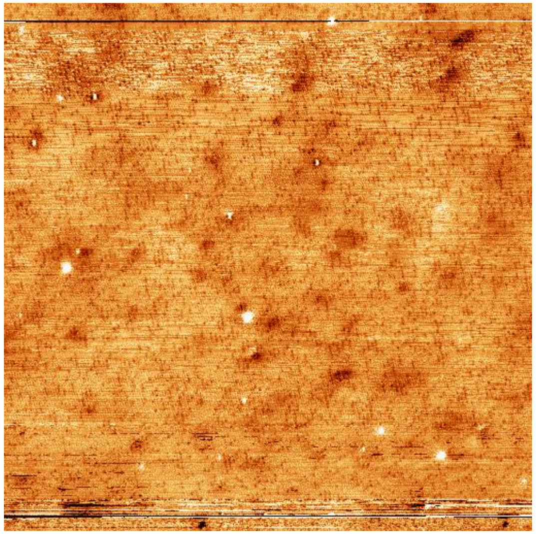
  width: 1 nm
  height: 50 pm

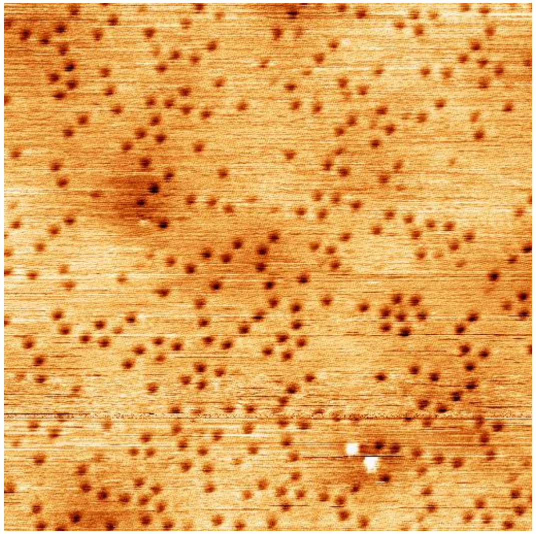
- small dark spots

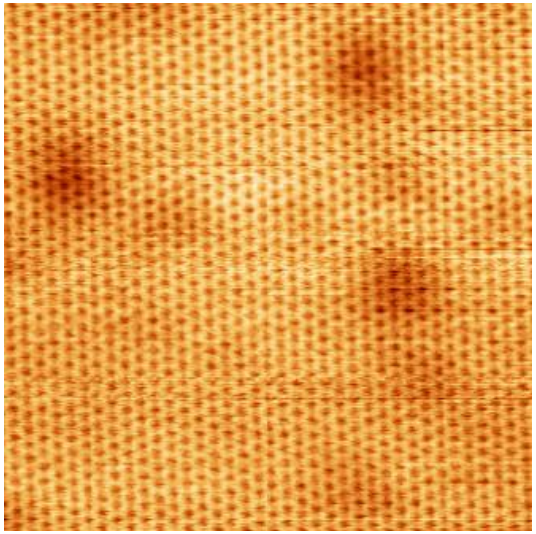
  width: 1.5 nm
  depth: 15-25 pm

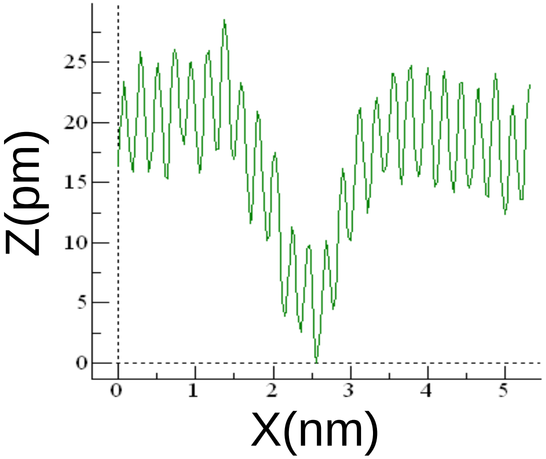


STM $T_H = 800ºC$


200 nm


50 nm


8 nm

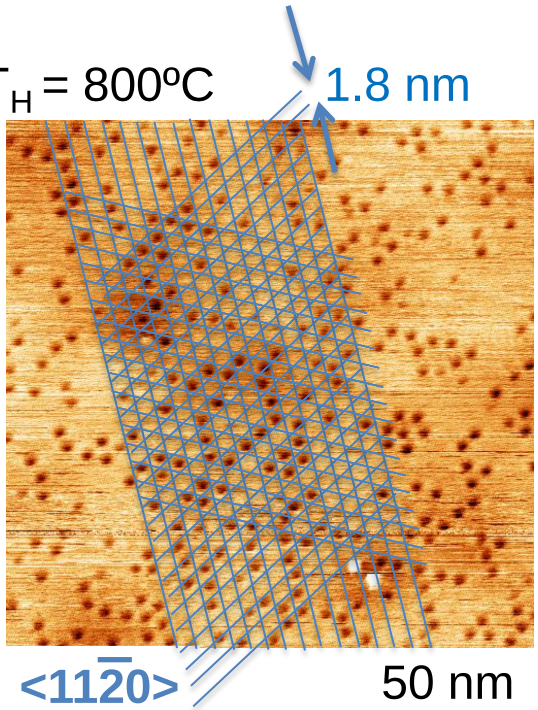


- small dark spots
- bright spots

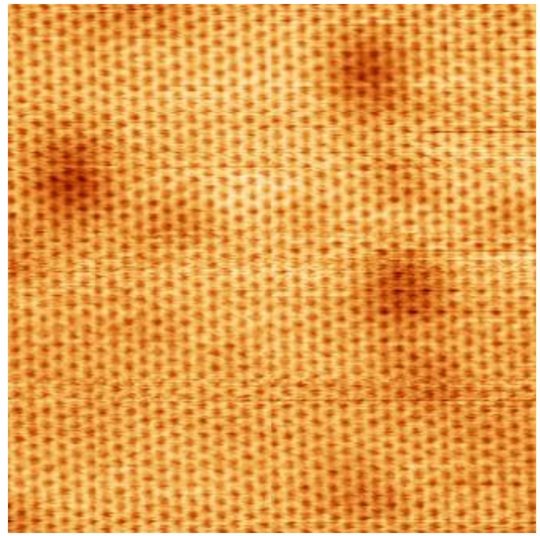
almost same morphology as $T_H = 600ºC$

Small dark spots

T_H = 800ºC 1.8 nm

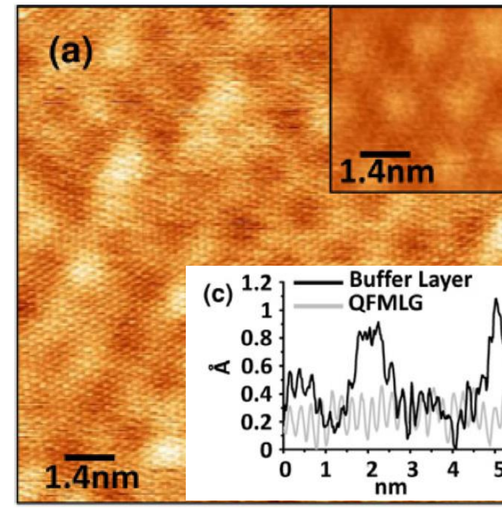


$\langle 11\bar{2}0 \rangle$ 50 nm



8 nm

STM of a buffer layer
corrugation – 60 pm



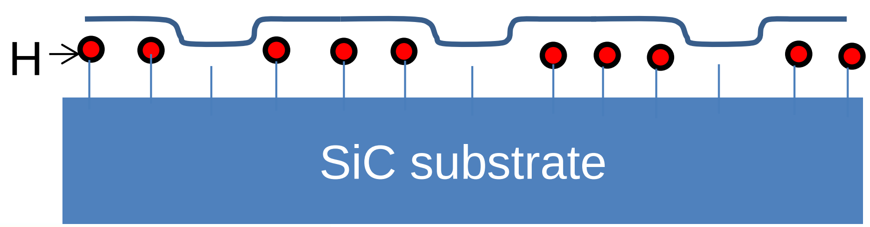
Goler, Carbon **51**, 249 (2013)

- width: 1.5 nm
- depth: 15-25 pm

- align along SiC$\langle 11\bar{2}0 \rangle$
- periodicity: 1.8 nm = SiC-6 × 6

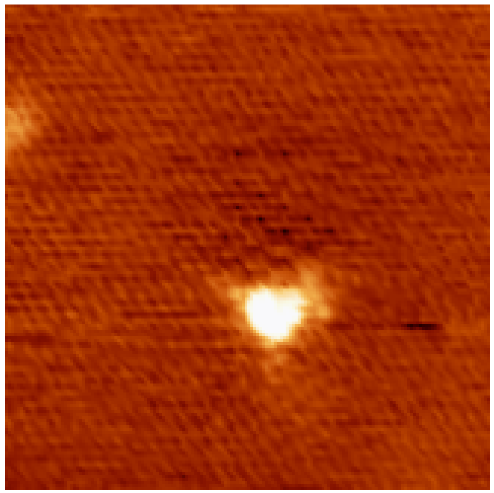
- honeycomb inside

incomplete H intercalation
– Si dangling bond at interface

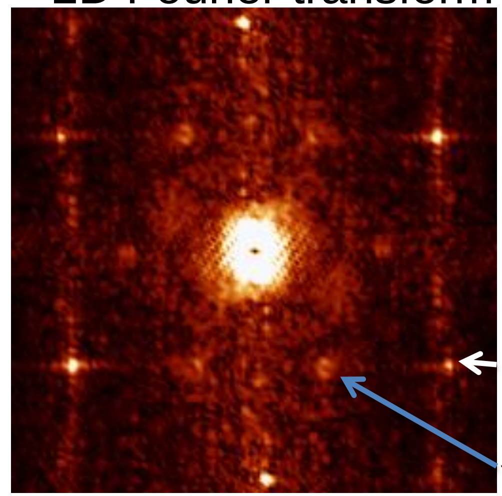


Bright spots

8 nm



2D Fourier transform

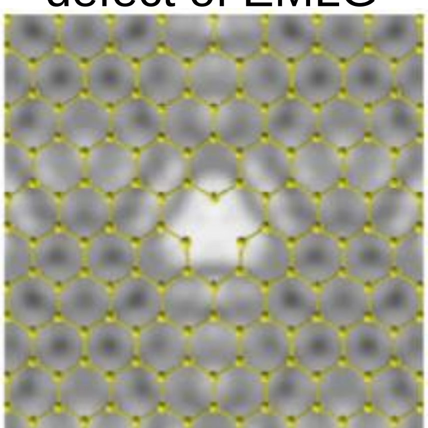


SiC(0001)-1 × 1

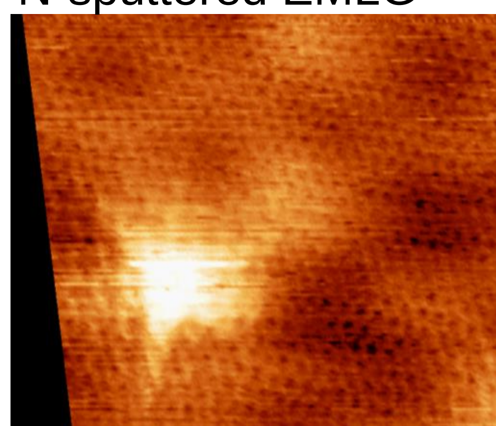
√3 × √3

electron scattering at defect

defect of EMLG

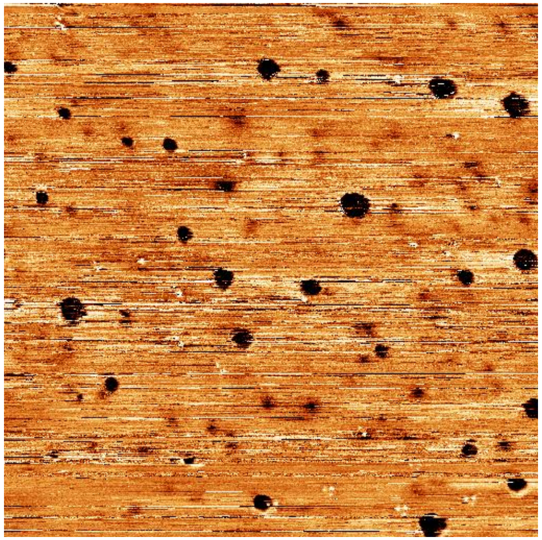


N-sputtered EMLG

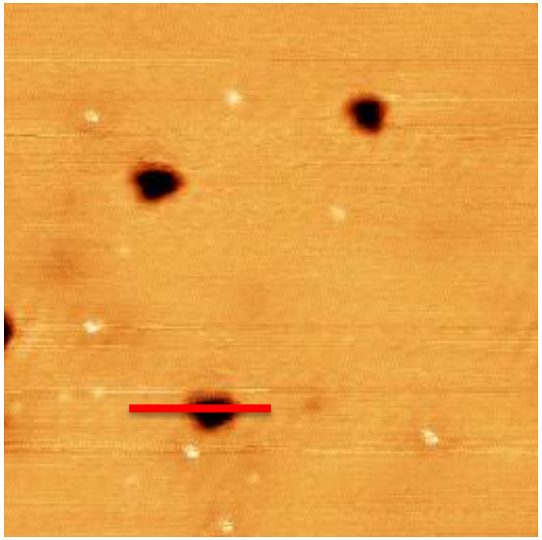


Rhim, Appl. Phys. Lett. **100**, 233119 (2012)

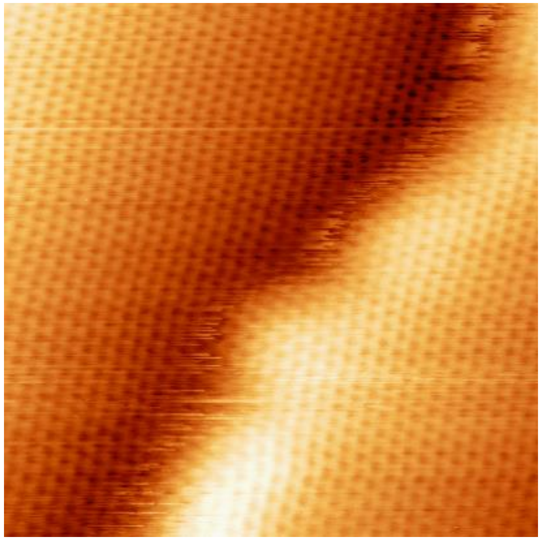
STM $T_H = 1000^oC$



200 nm

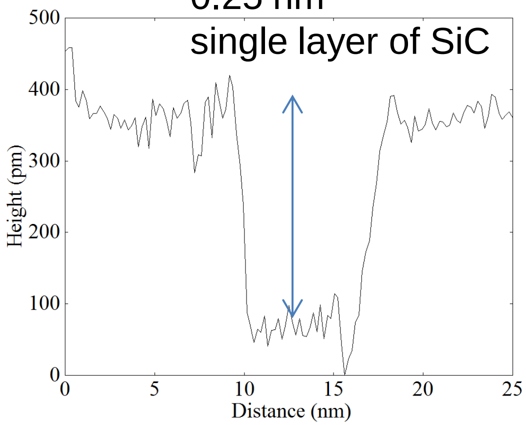


50 nm



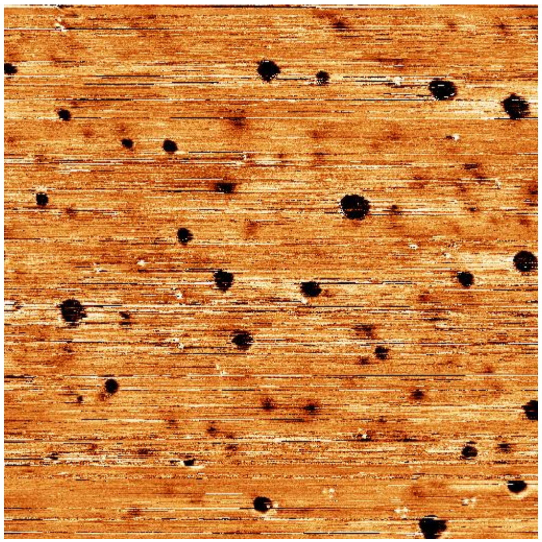
8 nm

0.25 nm ~
single layer of SiC

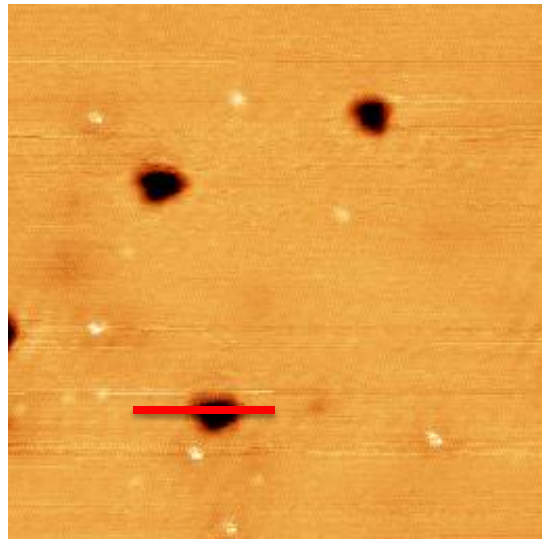


- large dark spots (width: 4-10 nm, depth: 0.25-0.3 nm)
- small dark spots (width: 1.5 nm, depth: 15-25 pm)
- bright spots

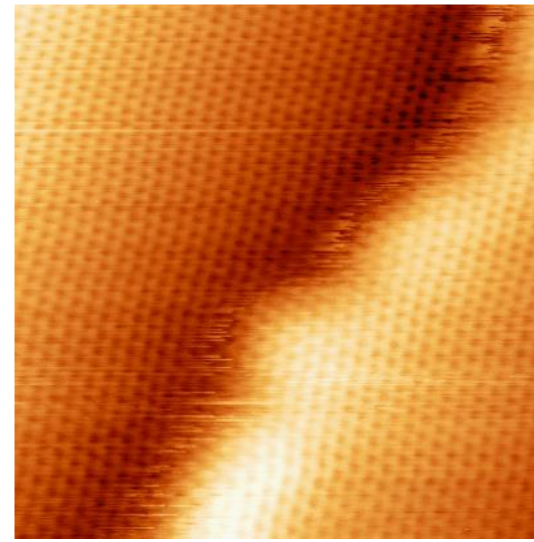
Large dark spots



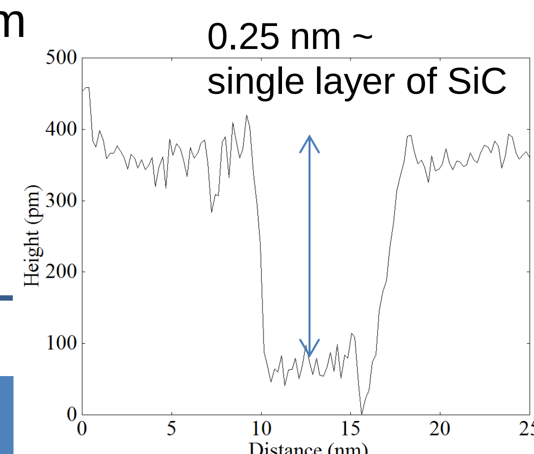
200 nm



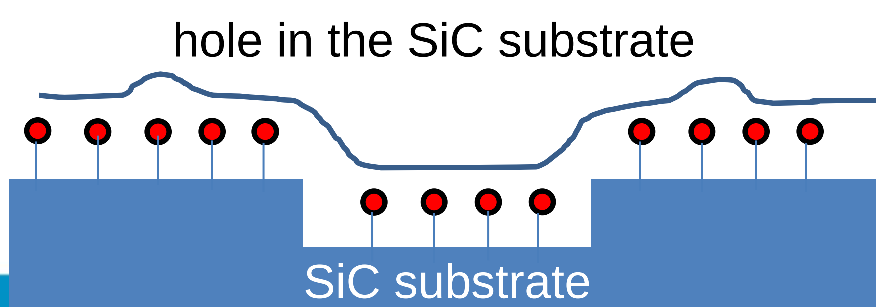
50 nm



8 nm



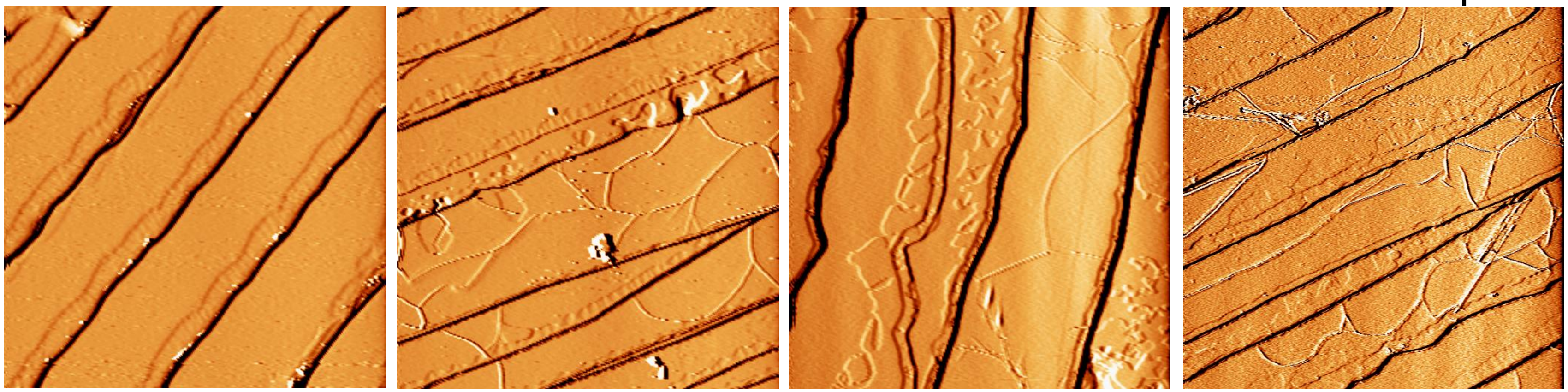
- width: 4-10 nm, depth: 0.25-0.3 nm
- random distribution
- honeycomb inside



etched at high T_H

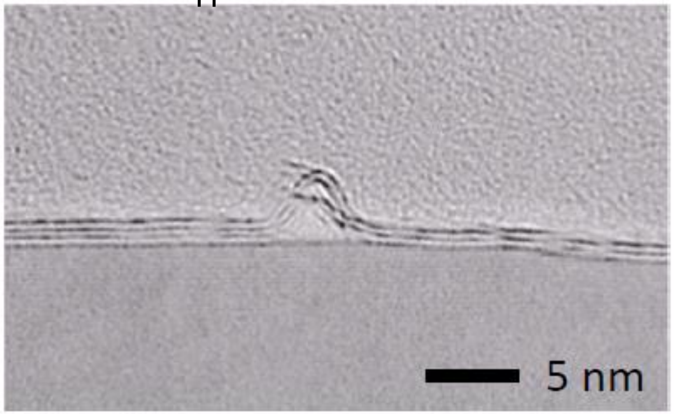
Wrinkles of graphene

AFM differentiated in horizontal axis 14 μm



T_H = 650ºC 800ºC 950ºC 1100ºC

TEM T_H = 1200ºC



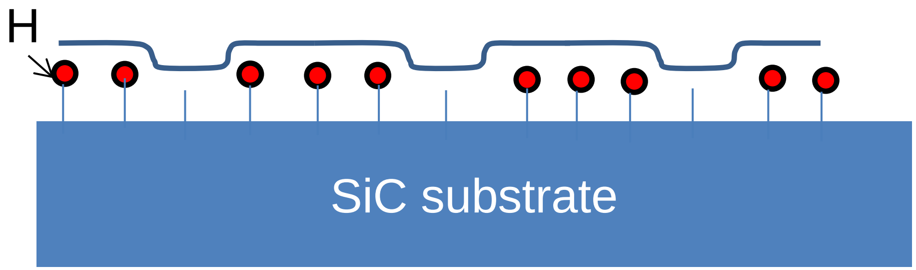
wrinkles appear at T_H > 800ºC

more frequently seen at T_H > 1100ºC

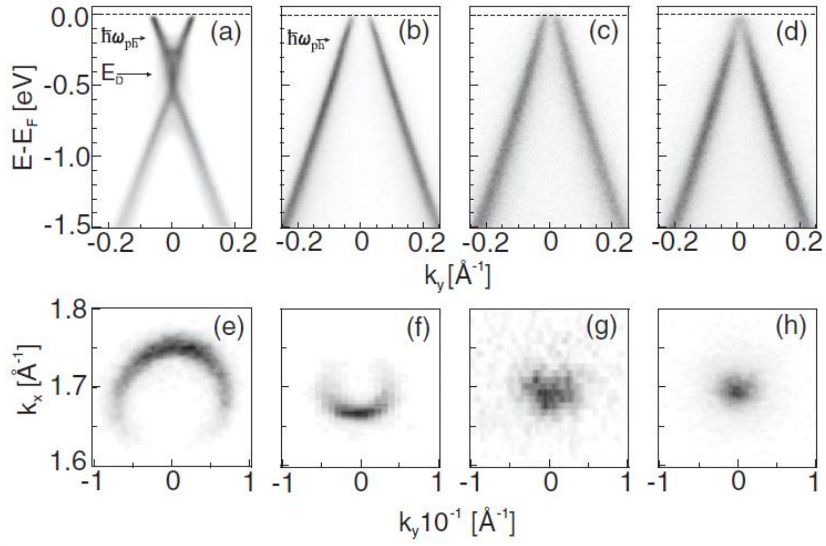
the difference in thermal expansion coefficients between graphene and SiC

Scattering in QFMLG

T_H = 600-800ºC



- small dark spots - Si dangling bonds due to incomplete H intercalation



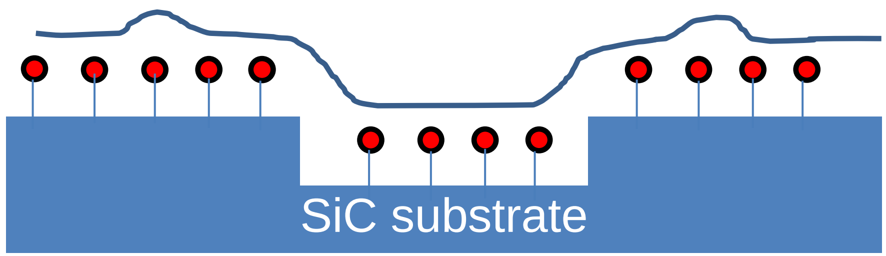
ARPES – H desorption from QFMLG

S. Forti, K. V. Emtsev, C. Coletti, A. A. Zakharov, C. Riedl, and U. Starke, Phys. Rev. B 84, 125449 (2011).

Si dangling bond donates charge to graphene and acts as a charged scattering center.

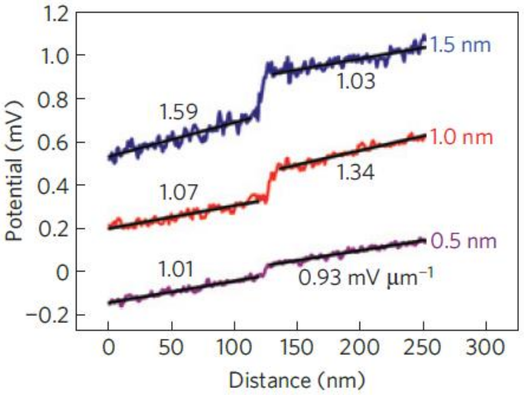
Scattering in QFMLG

$T_H = 1000^oC$



- dark spot – hole in SiC substrate
- wrinkle of graphene

EMLG resistance over SiC steps

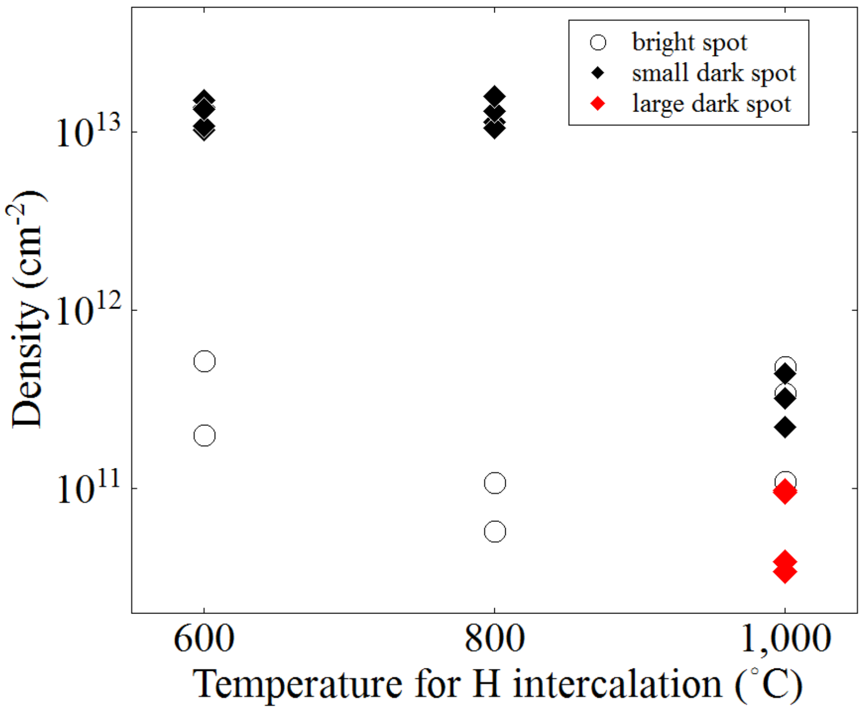


Scanning tunneling potentiometry
S. Ji, J. B. Hannon, R. M. Tromp, V. Perebeinos, J. Tersoff, and F. M. Ross, Nature Materials 11, 114 (2012)

- π-σ hybridization by curvature
- strain
- reduced doping from substrate

T. Low, V. Perebeinos, J. Tersoff, and Ph. Avouris, Phys. Rev. Lett. 108, 096601 (2012)

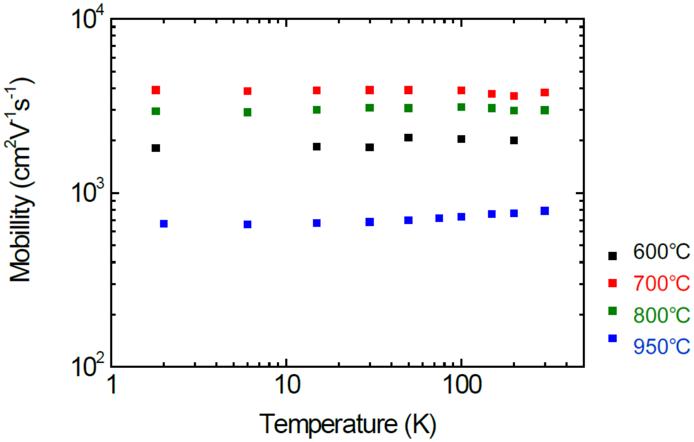
Scattering in QFMLG



As T_H increases from 600-800 to 1000ºC,

- small dark spot decreases.
  -more H intercalation
- large dark spot – hole in SiC substrate and wrinkles of graphene appear.
- bright spot – defect in graphene has the constant density < 0.016%.

The holes in SiC substrate and wrinkles of graphene are responsible for the lower mobility at 1000ºC.



S. Tanabe, Jpn. J. Appl. Phys. **53**, 04EN01 (2014).

Conclusion

- We investigated the morphology of QFMLG formed at several temperatures by H intercalation with STM, AFM, and TEM.

- We found that Si dangling bonds due to incomplete H intercalation at the graphene-substrate interface cause carrier scattering as charged impurities in QFMLG at T_H = 600 and 800ºC.

- At T_H = 1000ºC, holes in the SiC substrate and wrinkles of graphene appear and decrease the mobility of QFMLG, despite a better H intercalation.

- In order to obtain a higher mobility of QFMLG, we need to optimize the H intercalation condition to intercalate more H, below the temperature at which holes and wrinkles appear.

arXiv:1409.0457

CNRNANO PISA NEST

Thank you for your attention!

Funding:

Consiglio Nazionale delle Ricerche

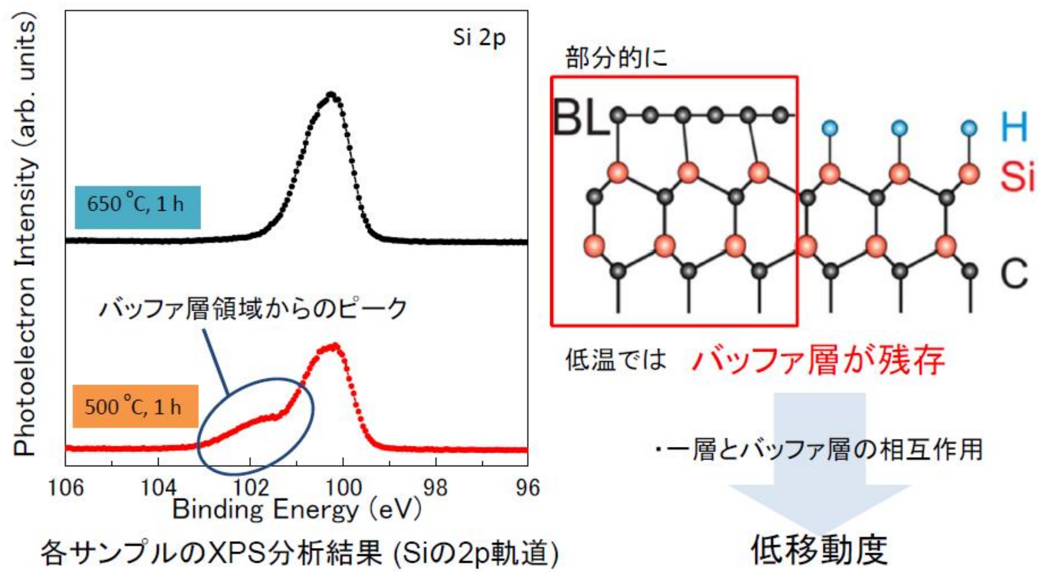
Japan Society for the Promotion of Science
日本学術振興会

Ministero degli Affari Esteri

GRAPHENE FLAGSHIP

National Enterprise for nanoScience and nanoTechnology
NEST

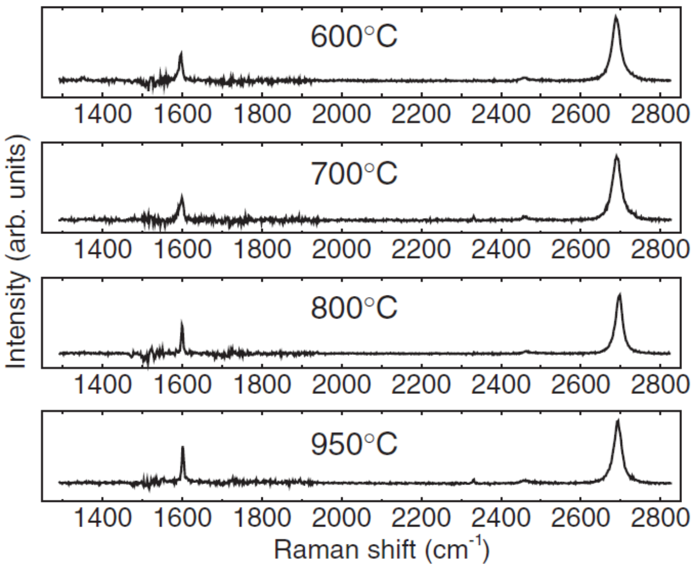
XPS

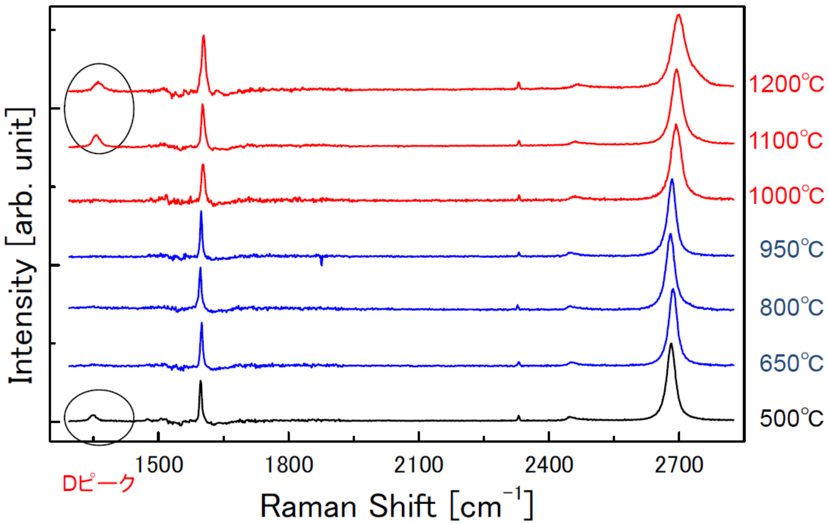


各サンプルのXPS分析結果 (Siの2p軌道)

The signal from buffer layer was obtained at T = 500C, but not at 650C.

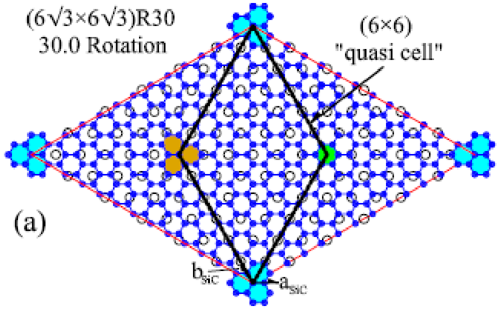
Raman





低温域および高温域ではDピークが見られた。
グラフェン中に欠陥が導入されることで低い移動度を示すと思われる。

D peak at 600C, 500C and > 1100C



A buffer layer (SiC(0001)1x1) has 1.22 x 10^{15} Si atoms cm^{-2}.

The density of small dark spots is <1.4 x 10^{13} cm^{-2} (~ 1% of Si atoms).