

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

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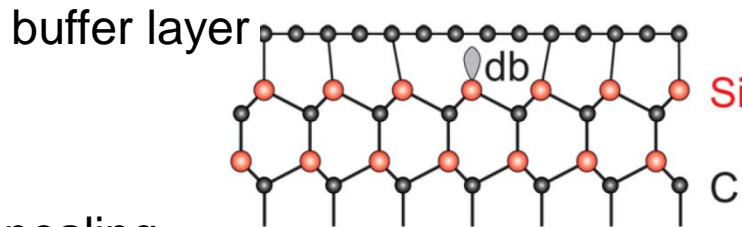
4 NTT Basic Research Laboratories, NTT Corporation, Japan

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Introduction

Graphene on silicon carbide (SiC) (0001)

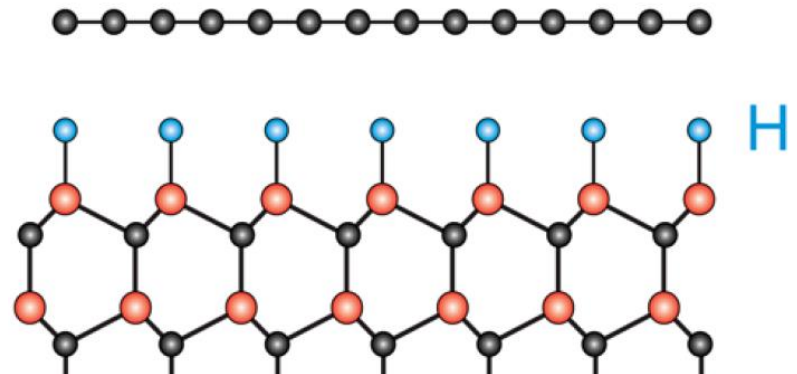
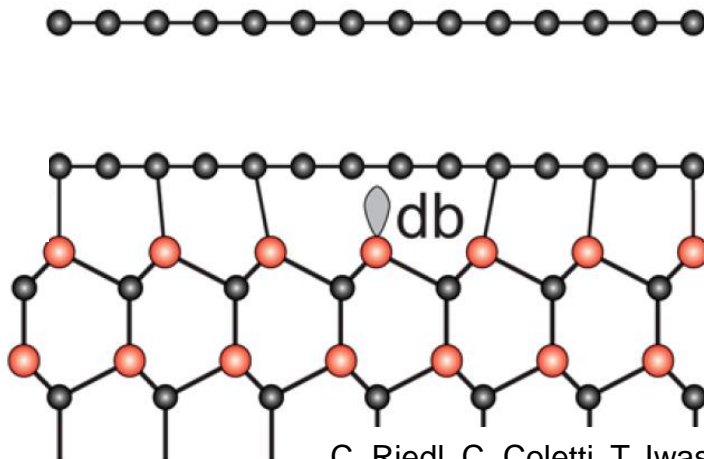


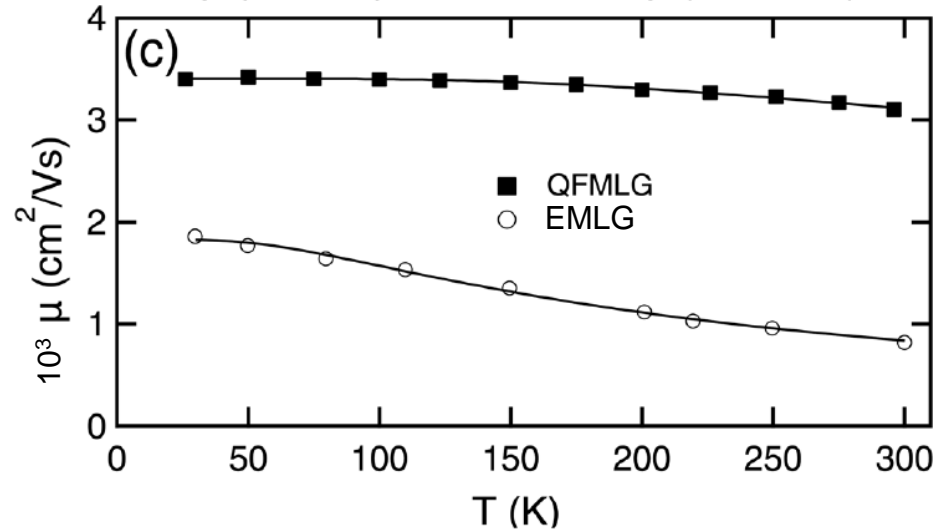
annealing

H intercalation

epitaxial monolayer
graphene (EMLG)

quasi-free-standing monolayer
graphene (QFMLG)

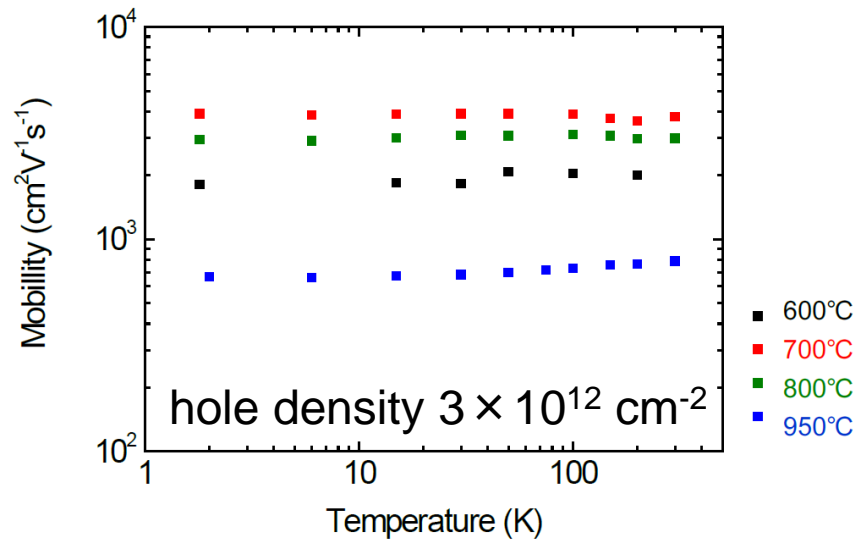




Carrier mobility of QFMLG: less temperature dependence
 less interaction between QFMLG and SiC substrate

However, mobility of QFMLG ($\sim 3000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) is 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).

- QFMLG mobility depends on T_H

T_H : temperature of substrate during H intercalation

- highest mobility by $T_H = 700^\circ\text{C}$

Purpose :

relationship between the morphology and transport property of QFMLG formed at different T_H

Experiment

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

cleaning anneal at 1500°C for 5 min in H₂ of 33 mbar

buffer layer growth

anneal at 1650°C for 5 min in Ar of 800 mbar

H intercalation

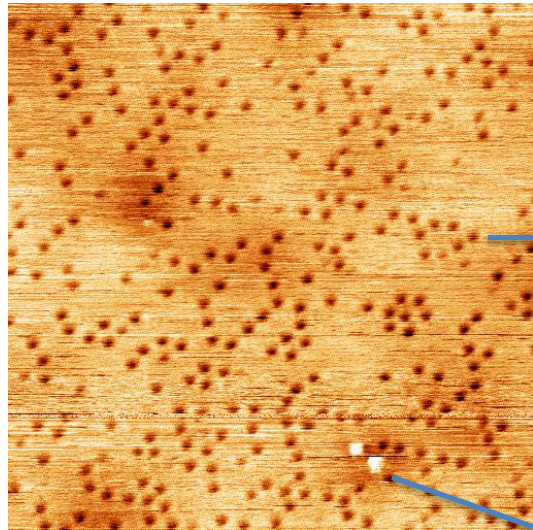
anneal at 600 - 1200°C for 1 hour in H₂ of 1013 mbar

characterization

- STM at RT and 6K in ultra-high vacuum (1×10^{-10} mbar)
- AFM
- TEM

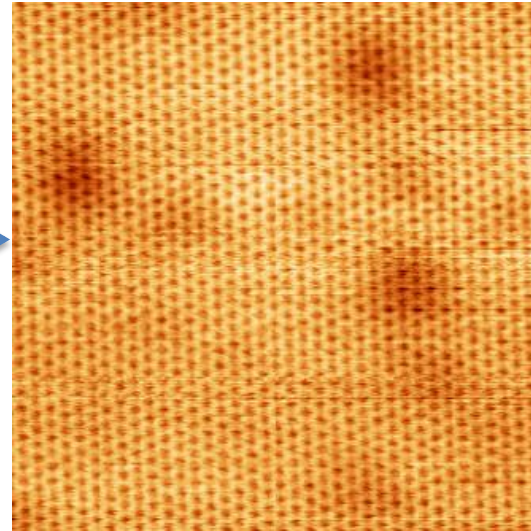
STM on QFMLG at RT

$T_H = 800^\circ\text{C}$ sample



50 nm

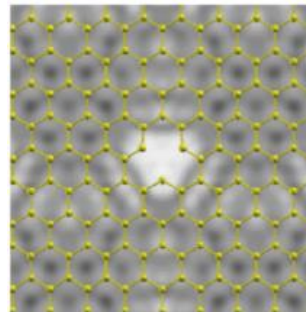
- dark site



width: 1.5 nm
depth: 20 pm

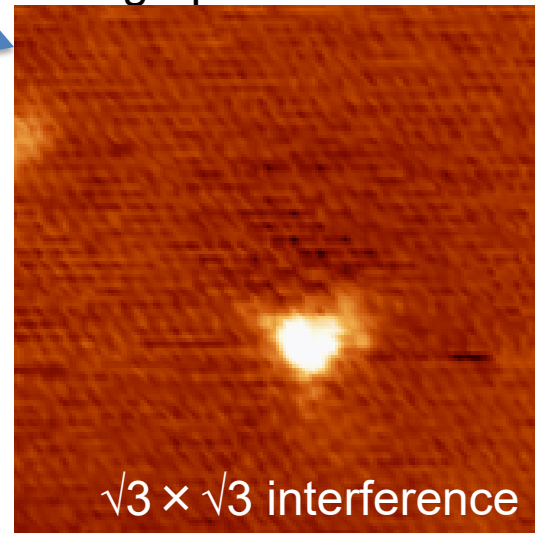
8 nm

simulated STM of defect



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

- graphene defect



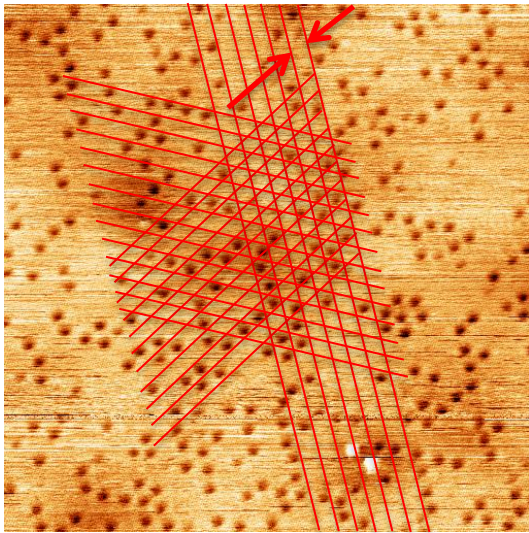
$\sqrt{3} \times \sqrt{3}$ interference

8 nm

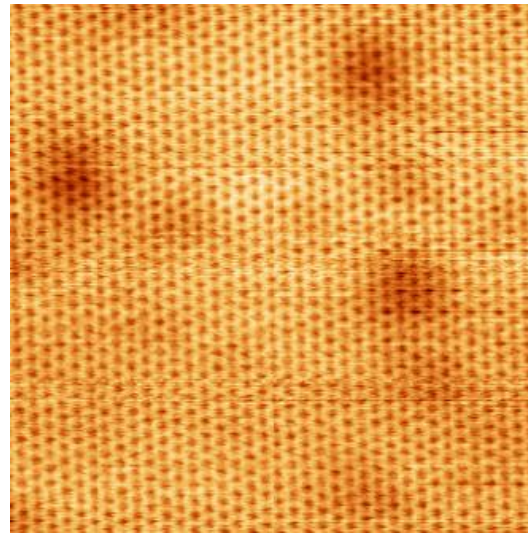
Si dangling bonds

$\langle 11\bar{2}0 \rangle$ directions
1.8 nm spacing

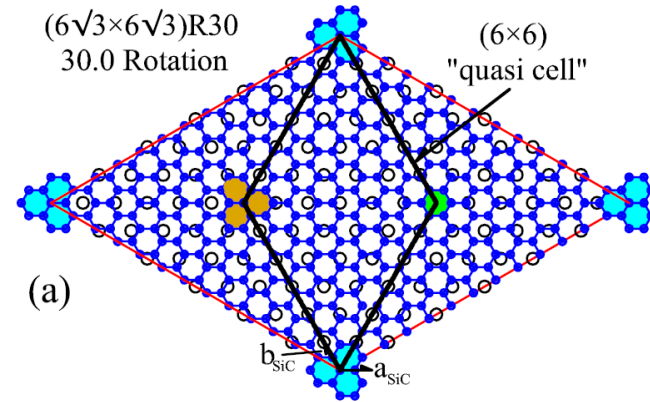
} ~ SiC 6×6 cell ~ moiré periodicity



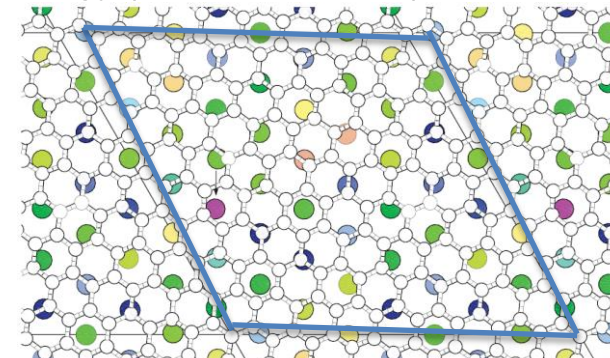
50 nm



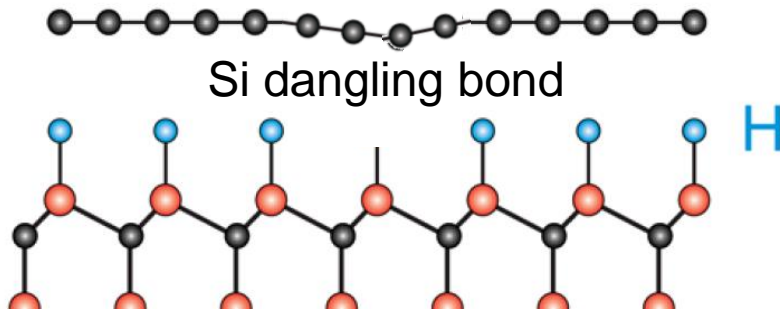
8 nm



spatial distribution of hydrogenation energy ($4\sqrt{3} \times 4\sqrt{3}$ model)

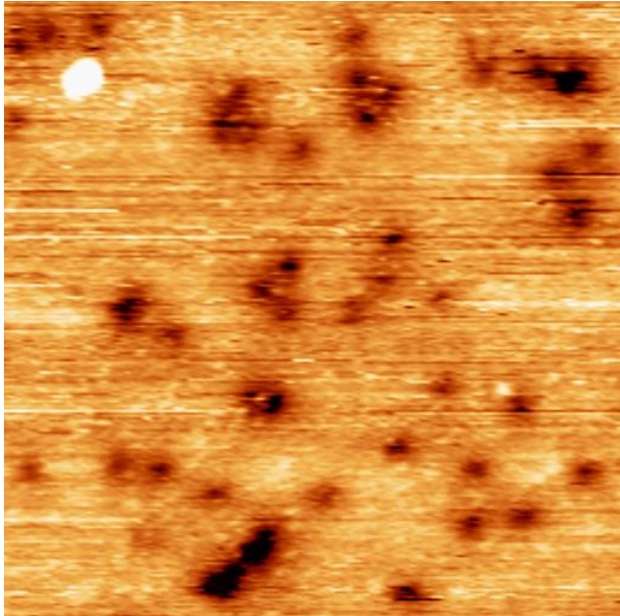


dark blue: the least favored H adsorption site

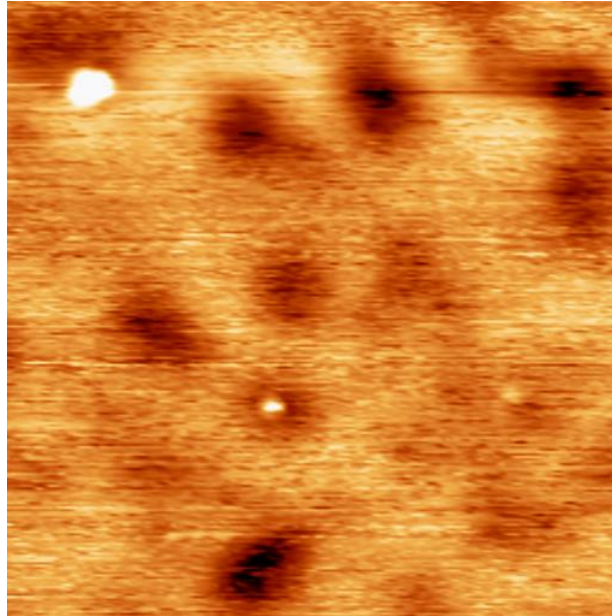


Si dangling bonds

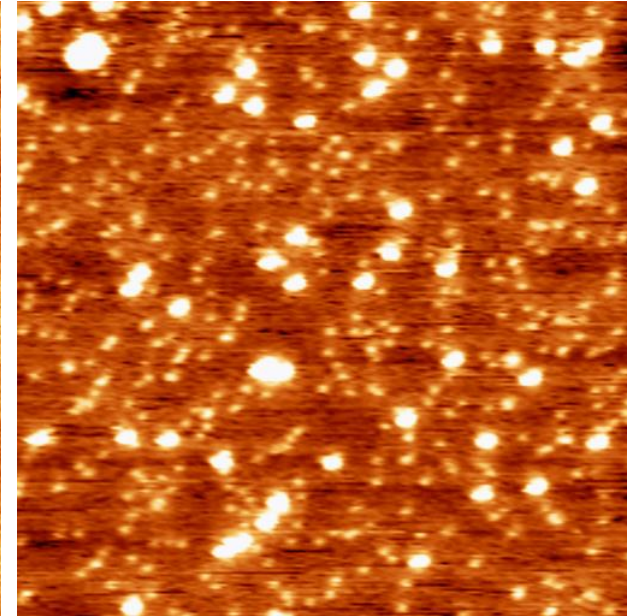
$V_s = -1.5V$ 0.1nA



-0.1V



1.5V

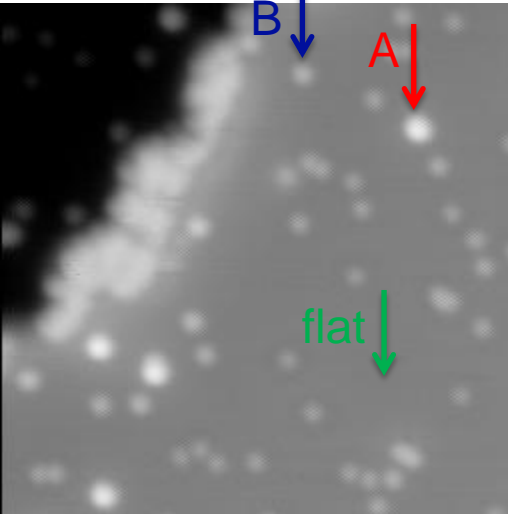


50nm

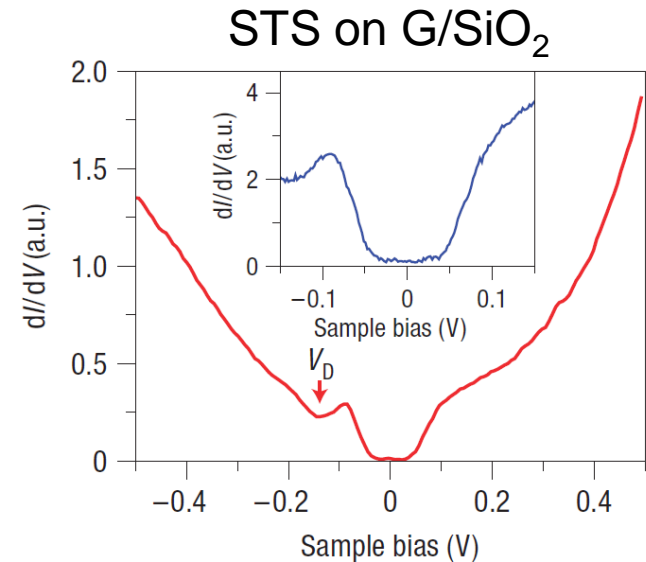
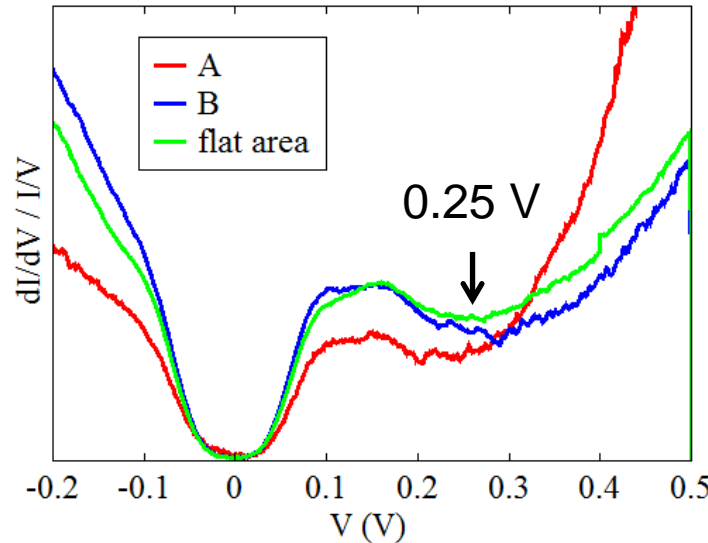
- contrast changes with V_s – electronic effect
- 2 different contrasts at 1.5V

Si dangling bonds

STM and STS at 6K



1.8V, 0.01nA, 20 nm

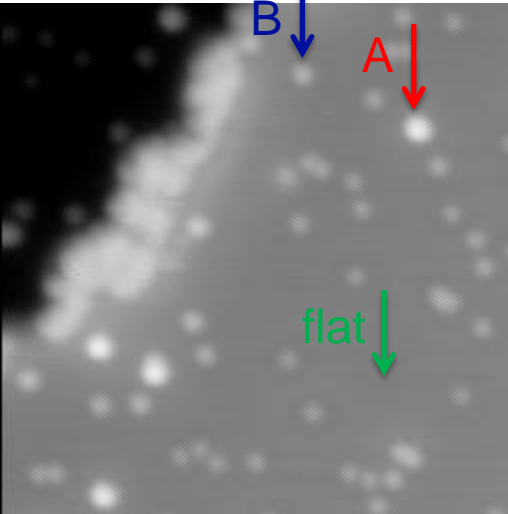


Y. Zhang, V. W. Brar, F. Wang, C. Girit, Y. Yayon, M. Panlasigui, A. Zettl, and M. F. Crommie, Nat. Phys. 4, 627 (2008).

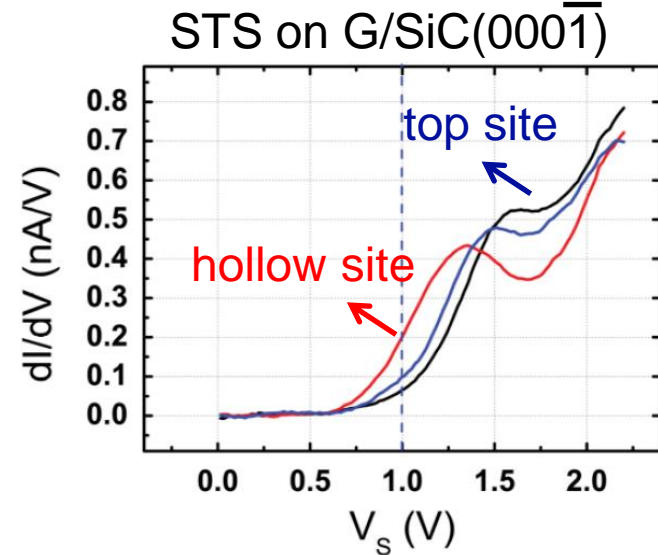
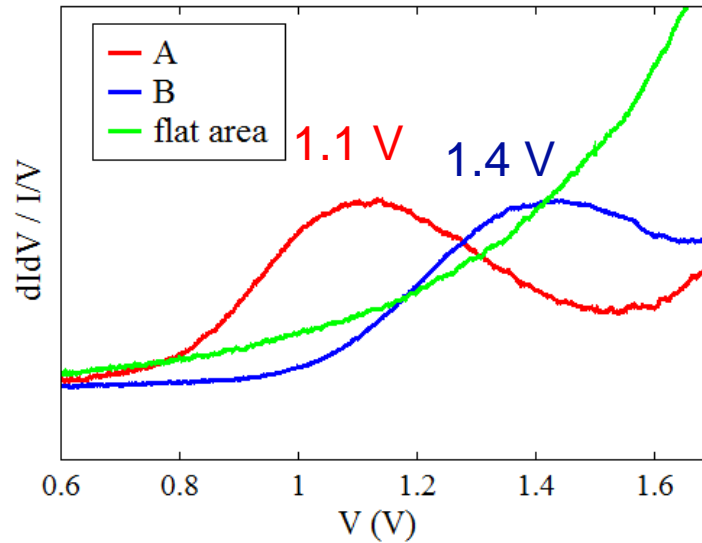
- p doping of graphene with shift of Dirac point by 0.25 V
- consistent with other measurements on QFMLG (ARPES, STS, Hall)
- spontaneous polarization of polar surface on SiC substrate

Si dangling bonds

STM and STS at 6K



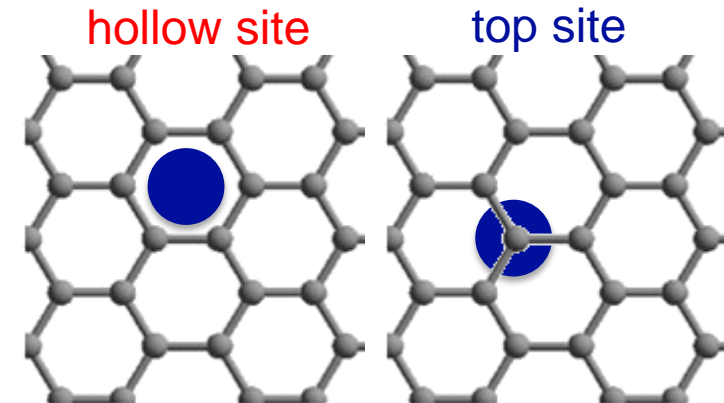
1.8V, 0.01nA, 20 nm



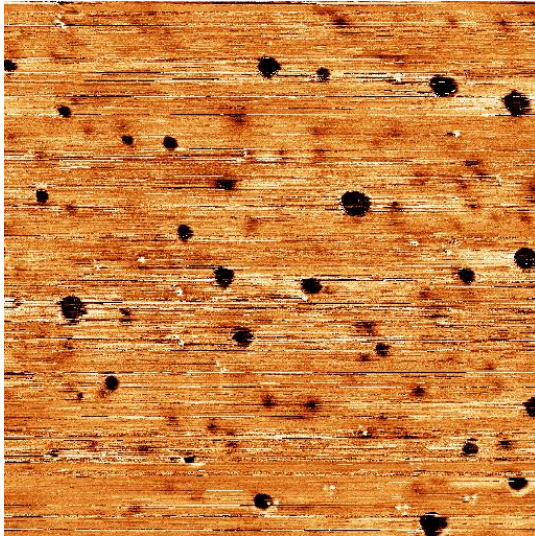
F. Hiebel, P. Mallet, J.-Y. Veullen, and L. Magaud, Phys. Rev. B 86, 205421 (2012).

- 2 types of Si dangling bonds at different graphene / Si stacking configurations

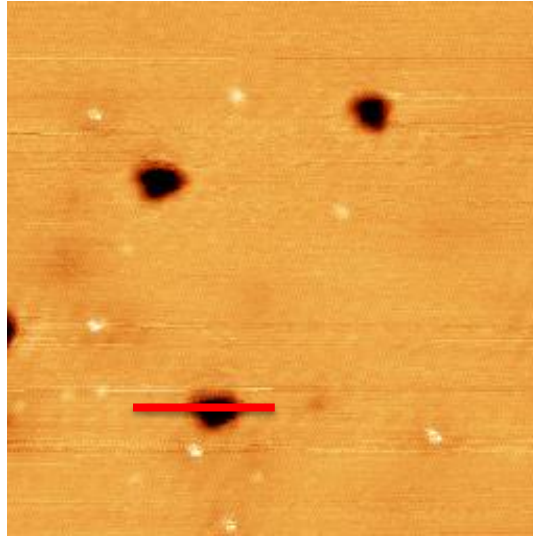
>>> DFT calculation of the electronic states in progress



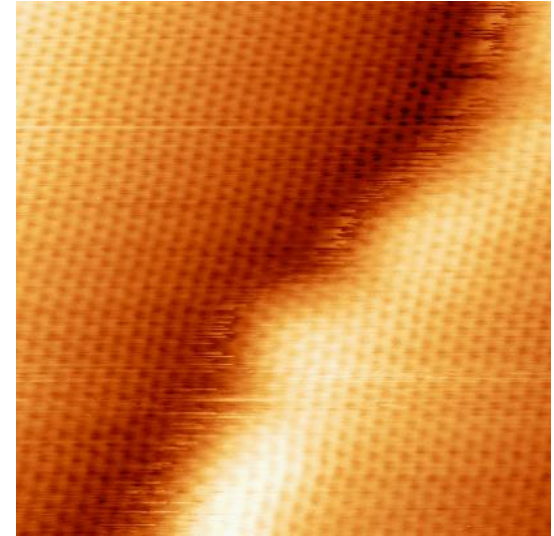
SiC holes: $T_H = 1000^\circ\text{C}$



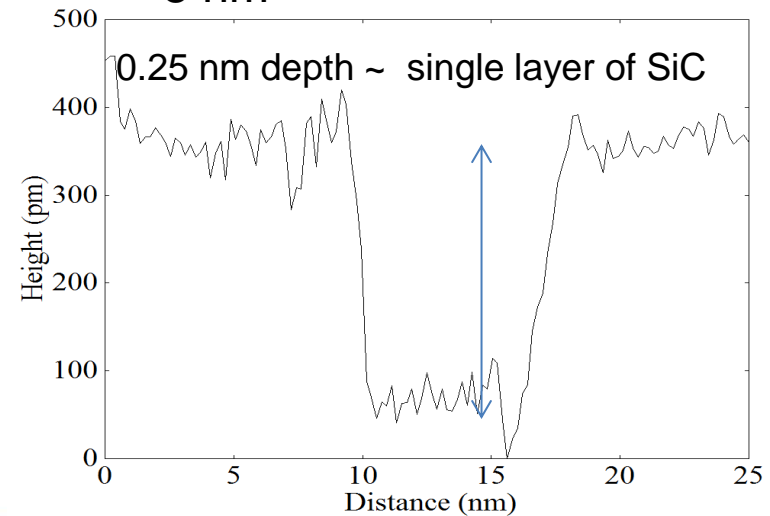
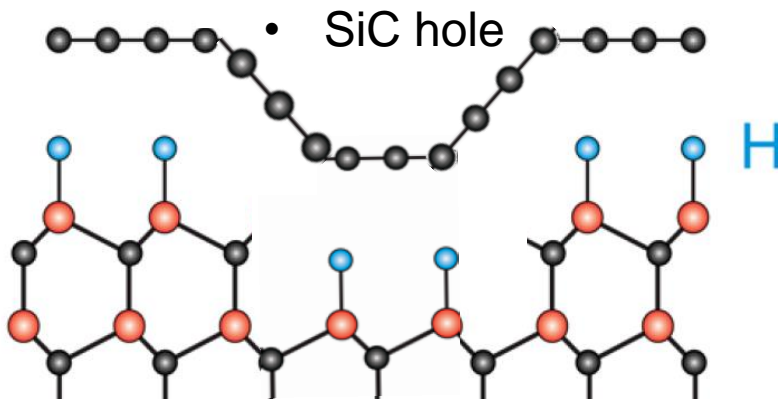
200 nm



50 nm



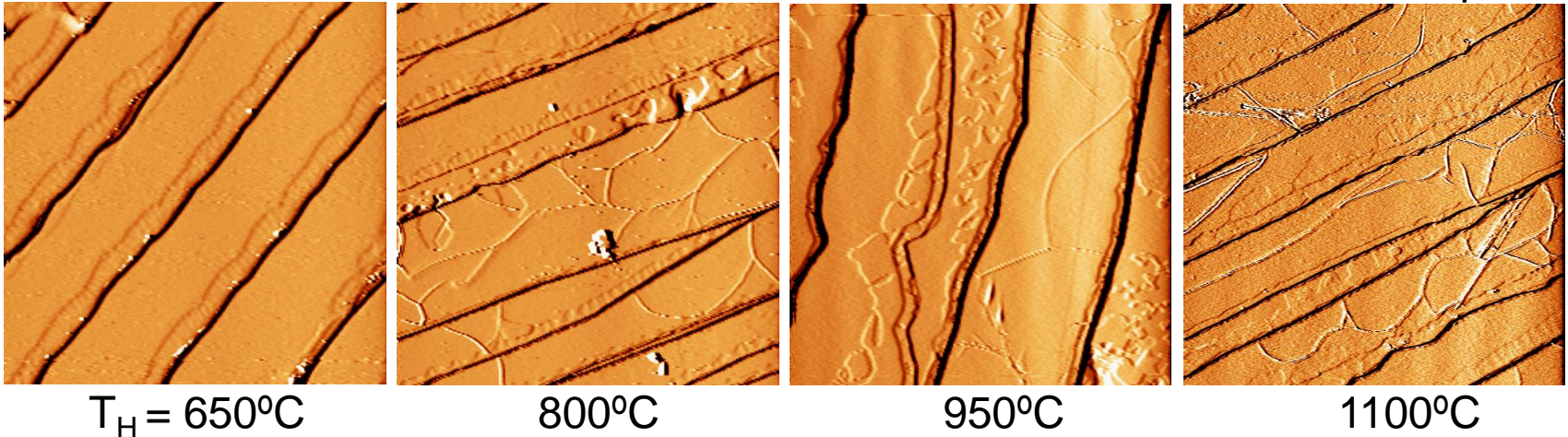
8 nm



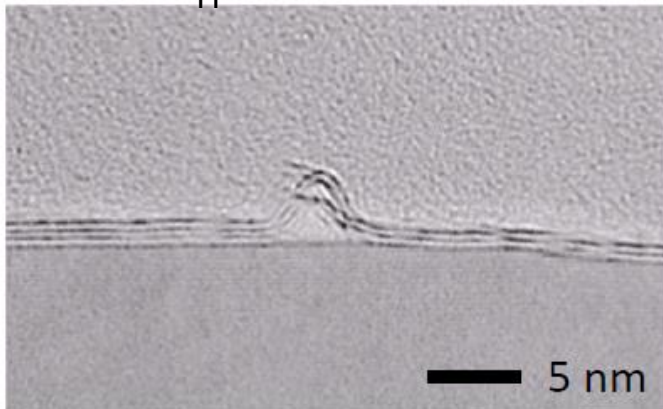
graphene wrinkles: $T_H = 1000^\circ\text{C}$

AFM

14 μm



TEM $T_H = 1200^\circ\text{C}$

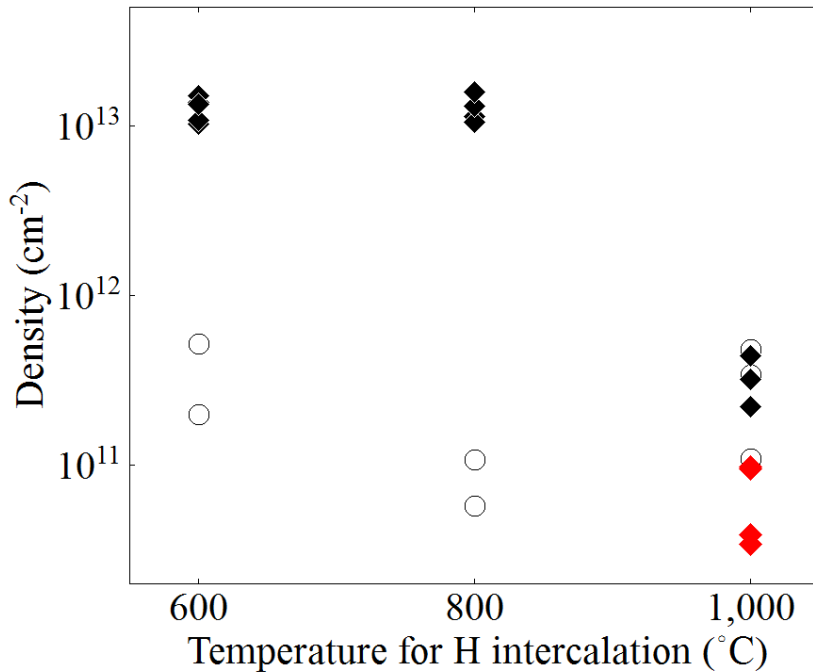


- graphene wrinkle

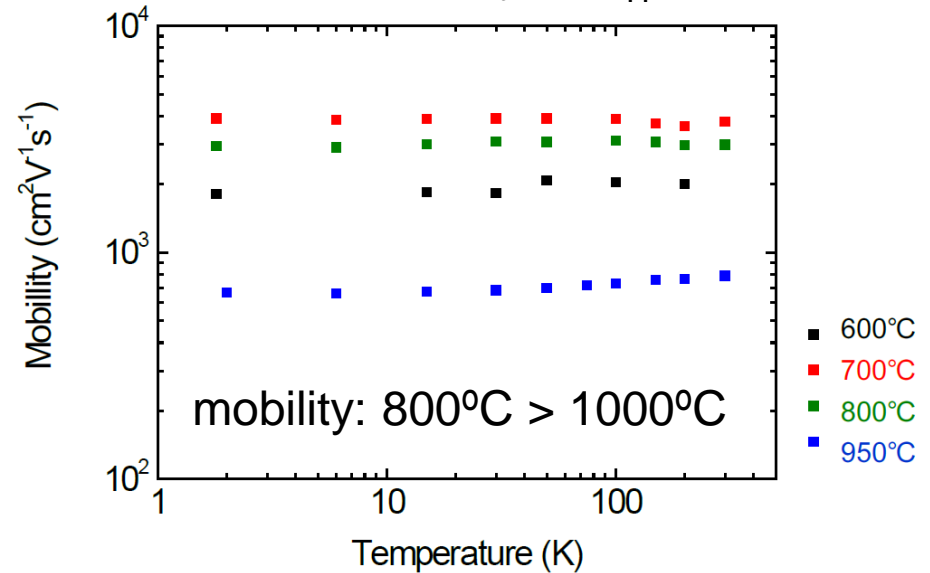
the difference in thermal expansion coefficients between graphene and SiC

Morphology and transport

densities of features on QFMLG



mobility vs T_H



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

- graphene defect ----- constant with T_H • influence on mobility
- ◆ Si dangling bonds ----- decrease with T_H ----- small increase
- ◆ SiC hole (and G wrinkle) ----- increase with T_H ----- large decrease

Murata, Appl. Phys. Lett. 105, 221604 (2014)

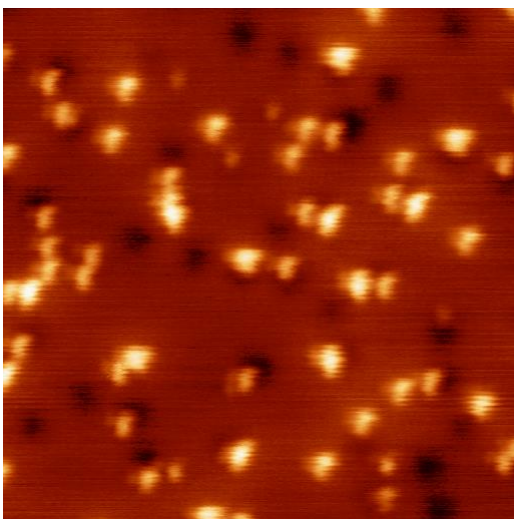
Conclusion

- We investigated the morphology of QFMLG formed at different temperatures for H intercalation with STM, AFM, and TEM.
- graphene defect and Si dangling bond
- additionally, SiC holes and graphene wrinkle on $T_H = 1000^\circ\text{C}$ sample
- 2 Si sites with different electronic states. The difference may be attributed to different local graphene / Si stacking configurations.
- In order to obtain a high mobility QFMLG, we need to optimize H intercalation condition to intercalate more H, below the temperature at which SiC holes and graphene wrinkles appear.

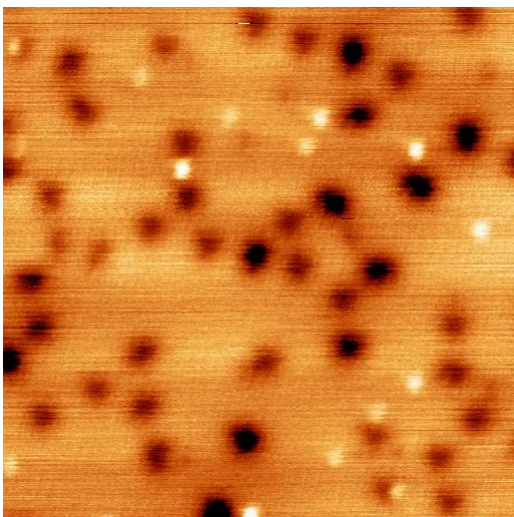
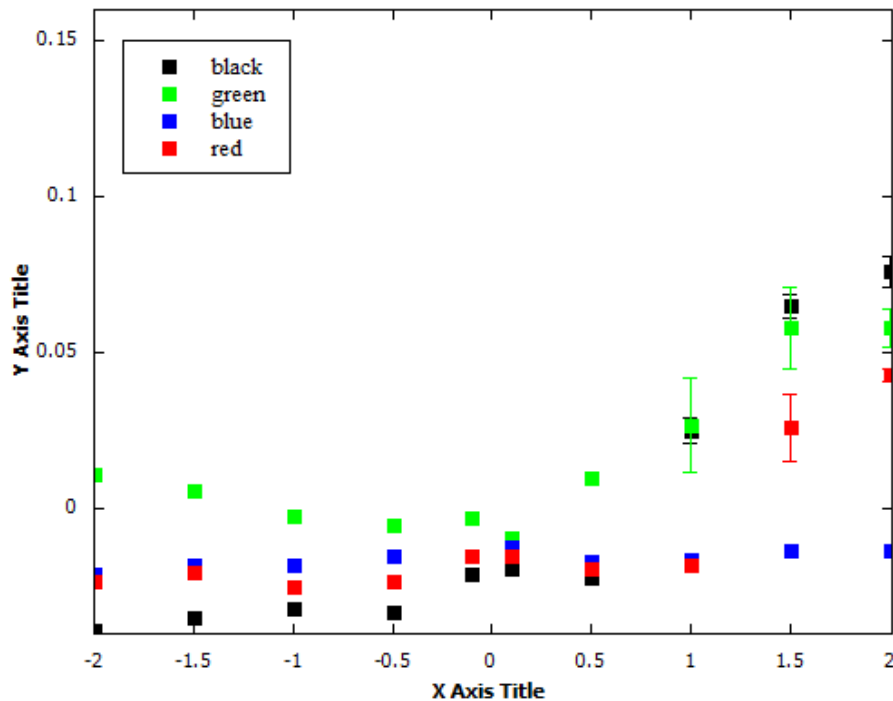


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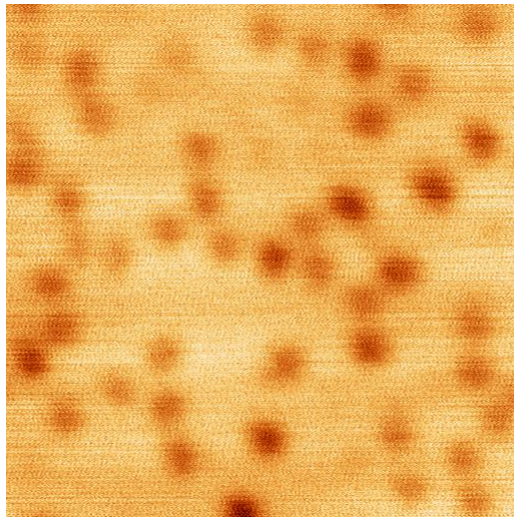
NEST



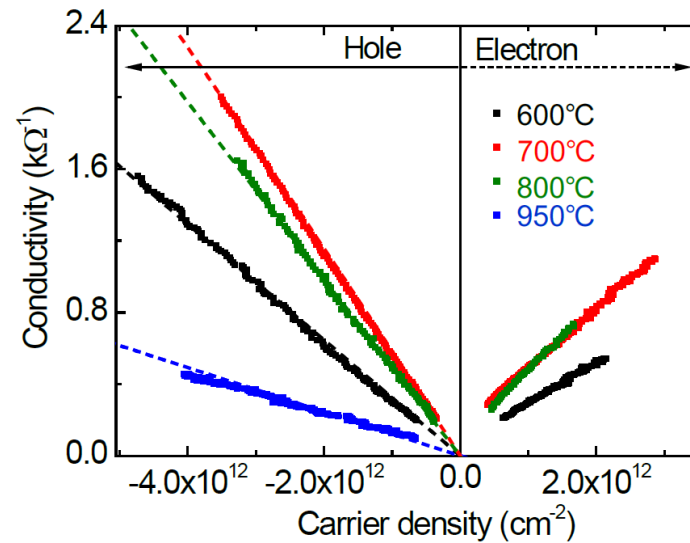
2V 0.1nA 20nm



-2V 0.1nA 20nm



0.1V 0.1nA 20nm



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

conductivity – carrier density

- linear for $T_H = 600-800^\circ\text{C}$
- charged impurity
- sublinear for $T_H = 950^\circ\text{C}$
- additional scattering by defect