分野２　CMSI

実空間密度汎関数法による ナノ物質の第一原理 シミュレーション

実空間密度汎関数法コード – *RSDFT* –

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- Kenichi Koizumi (The University of Tokyo)

Computer Science
Material Science

- 筑波大（櫻井研）二村保徳（櫻井-杉浦の固有値解法）
- 東大（押山研） 内田和之（二層グラフェン）
- 東大（押山研） 澤田啓介（SiCステップ表面）

SC11

ACM Gordon Bell Prize

Peak Performance

Yukihiro Hasegawa, Junichi Iwata, Miwako Tsuji, Daisuke Takahashi, Atsushi Oshiyama, Kazuo Minami, Taisuke Boku, Fumiyoshi Shoji, Atsuya Uno, Motoyoshi Kurokawa, Hikaru Inoue, Ikuo Miyoshi, Mitsuo Yokokawa

First-Principles Calculation of Electronic States of a Silicon Nanowire with 100,000 Atoms on the K Computer

Scott Lathrop
SC11 Conference Chair

Thom H. Dunning, Jr.
Gordon Bell Chair

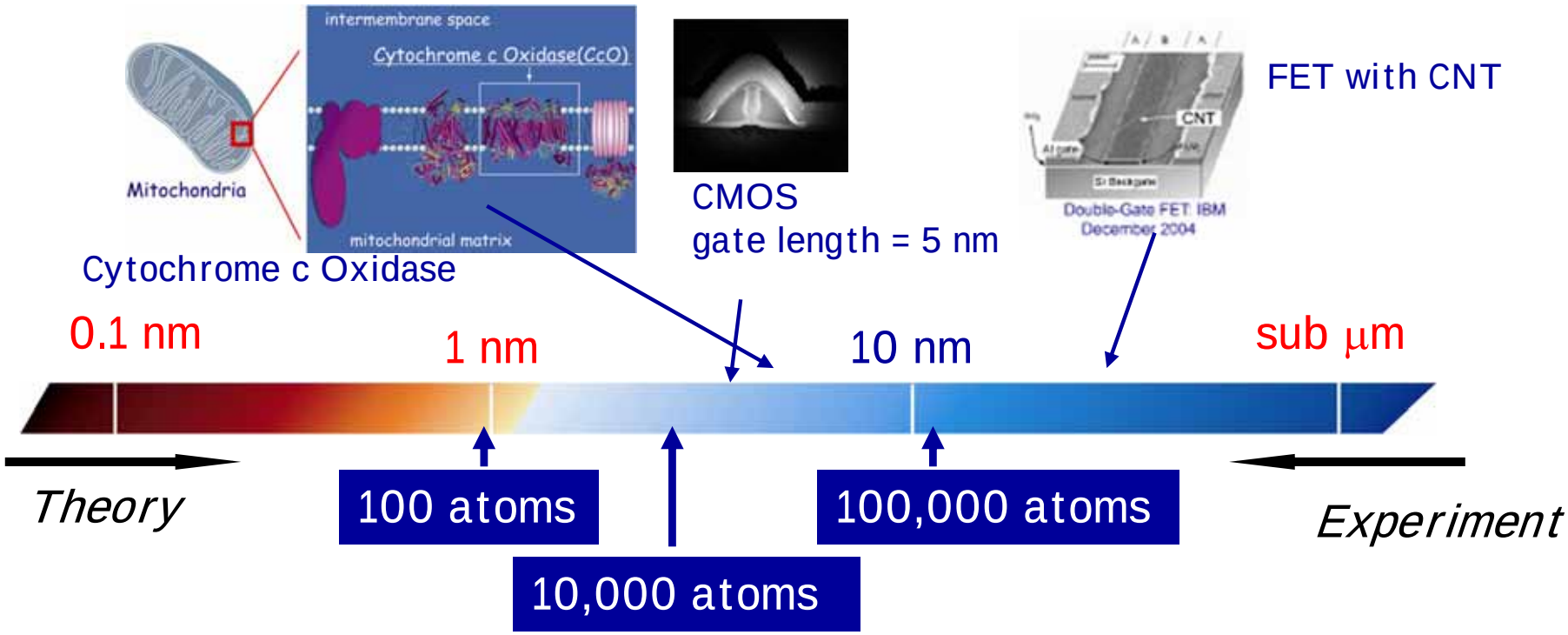
内容

- 第一原理電子状態計算
  - 密度汎関数法
  - RSDFT
- Siナノワイヤ
- 櫻井-杉浦法のバンド構造計算への応用（筑波大 二村保徳）
- 捩じれた二層グラフェン（東大 内田和之）
- SiCステップ表面（東大 澤田啓介）
- RSDFT-CPMDの実装（東大 小泉健一）

FIRST-PRINCIPLES CALCULATIONS

Density Functional Theory

LARGE-SCALE FIRST-PRINCIPLES CALCULATIONS IN NANO WORLD



Large-scale DFT calculations and experiments meet together in Nano World !

Challenge: 10,000 ~ 100,000-atom calculations overcoming N^3 scaling to reveal nano-scale world!

DENSITY FUNCTIONAL THEORY

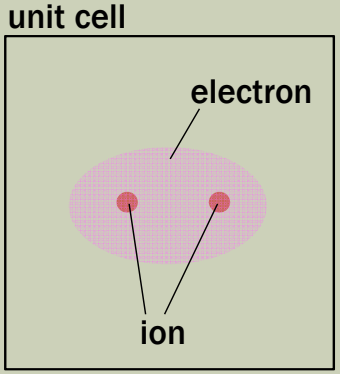
Energy functional

$$E[\{\phi_i\}] = \sum_{i=1}^{N} \int d\mathbf{r} \phi_i^*(\mathbf{r}) \left(-\frac{1}{2} \nabla^2 \phi_i(\mathbf{r}) \right)$$ 運動エネルギー

$$+ \int d\mathbf{r} \rho(\mathbf{r}) v_{ion}(\mathbf{r})$$ 電子–イオン相互作用

$$+ \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \frac{\rho(\mathbf{r}) \rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}$$ 電子間クーロン相互作用

$$+ E_{XC}[\rho]$$ その他量子力学的効果（交換相関効果）



Electron density

$$\rho(\mathbf{r}) = \sum_{i=1}^{N} |\phi_i(\mathbf{r})|^2$$

P. Hohenberg and W. Kohn, Phys. Rev. 136 (1964) B864.

W. Kohn and L. J. Sham, Phys. Rev. 140 (1965) A1133.

KOHN-SHAM EQUATION

汎関数最小化

$$\frac{\delta E[\phi]}{\delta \phi_i^*(\mathbf{r})} = 0 \qquad \left(\text{constraint} \quad \int d\mathbf{r} \phi_i^*(\mathbf{r}) \phi_j(\mathbf{r}) = \delta_{ij} \right)$$

➡ 系の最安定状態（基底状態）

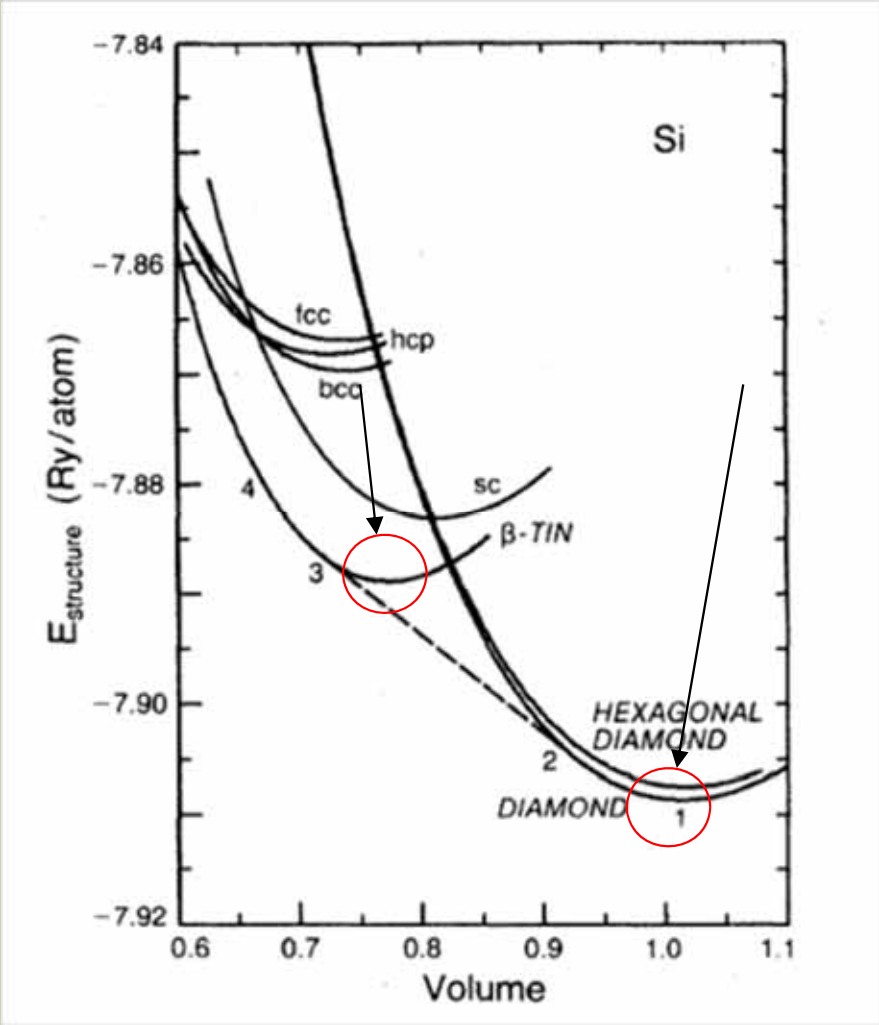
変分方程式：**Kohn-Sham方程式**　W. Kohn and L. J. Sham, Phys. Rev. 140 (1965) A1133.

$$\left(-\frac{1}{2}\nabla^2 + v_{KS}(\mathbf{r}) \right) \phi_i(\mathbf{r}) = \varepsilon_i \phi_i(\mathbf{r})$$

電子密度 $\rho(\mathbf{r}) = \sum_{i=1}^{N} |\phi_i(\mathbf{r})|^2$ ⟶ $v_{KS}(\mathbf{r}) = \int d\mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + \frac{\delta E_{XC}}{\delta \rho(\mathbf{r})} + v_{ion}(\mathbf{r})$

ポテンシャル

密度汎関数法の威力



M. T. Yin and M. L. Cohen, Phys. Rev. B26, 5668 (1982).

単純な近似で

$$E_{XC}[\rho] = E_X[\rho] + E_C[\rho]$$

局所密度近似(LDA)による交換汎関数

$$E_X[\rho] = -C_X \int d\mathbf{r} \rho^{5/3}(\mathbf{r})$$

$$v_X[\rho](\mathbf{r}) = \frac{\delta E_X[\rho]}{\delta \rho(\mathbf{r})} = -\frac{5}{3} C_X \rho^{1/3}(\mathbf{r})$$

高い定量性

Si（ダイヤモンド構造）

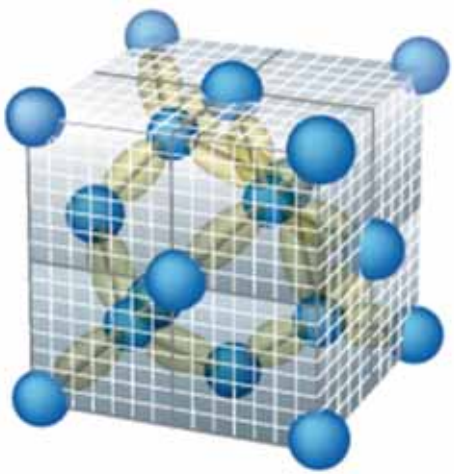
	DFT計算	実験値
格子定数(Å)	5.37	5.41
体積弾性率(Mb)	0.977	0.988

DFT CALCULATION IN REAL-SPACE GRID METHOD

RSDFT

REAL-SPACE FINITE-DIFFERENCE PSEUDOPOTENTIAL METHOD

FFT free



$$\left(-\frac{1}{2}\nabla^2+\hat{v}_{ion}+\int d\mathbf{r}'\frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}+\frac{\delta E_{XC}[\rho]}{\delta\rho(\mathbf{r})}\right)\phi_j(\mathbf{r})=\varepsilon_j\phi_j(\mathbf{r})$$

⟹ **Kohn-Sham equation is solved in discretized space**

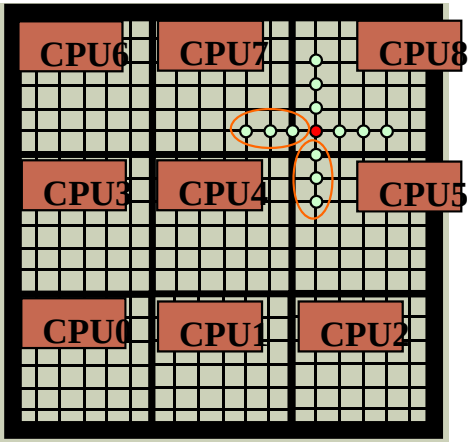
J. R. Chelikowsky *et al.*, Phys. Rev. B50, 11355 (1994).
J.-I. Iwata *et al.*, J. Comp. Phys. 229, 2339 (2010).

- Derivatives → (higher-order) finite difference $\frac{\partial^2}{\partial x^2}\phi(\mathbf{r})\approx\sum_{m=-6}^{6}C_m\phi(x+m\Delta,y,z)$
- Integrals → summation over grid points $\int\phi_m^*(\mathbf{r})\phi_n(\mathbf{r})d\mathbf{r}\approx\sum_{i=1}^{N_{grid}}\phi_m^*(\mathbf{r}_i)\phi_n(\mathbf{r}_i)\Delta V$
- Ionic potentials → Pseudopotentials $\hat{v}_{ion}=v_{local}(\mathbf{r})+\sum_{a,l,m}|\beta_{alm}\rangle\langle\beta_{alm}|$

N. Troullier & J. L. Martins
Phys. Rev. B 34, 1993 (1991)

GRID, BAND, k, SPIN PARALLELIZATION

- MPI (Message-Passing Interface) library
  - MPI_ISEND, MPI_IRECV → finite-difference calc.
  - MPI_ALLREDUCE → global summation
- OpenMP
  - Further grid parallelization (within each CPU) is performed by thread parallelization



Example of CPU allocation for 4-grid, 2-orbital, 2-k, 1-spin parallelization

Spin1

BZ1

Orbital1

Grid0 (CPU0)	*Grid2* (CPU2)
Grid1 (CPU1)	*Grid3* (CPU3)

Orbital2

Grid0 (CPU4)	*Grid2* (CPU6)
Grid1 (CPU5)	*Grid3* (CPU7)

BZ2

Orbital1

Grid0 (CPU8)	*Grid2* (CPU10)
Grid1 (CPU9)	*Grid3* (CPU11)

Orbital2

Grid0 (CPU12)	*Grid2* (CPU14)
Grid1 (CPU13)	*Grid3* (CPU16)

計算上の問題点

SiNW110 (20nm diameter)
6309 atoms
3600 nodes

ML=660 x 660 x 12
MB=13440
4 sample k points

(second)

	SCF	DIAG	GS	CG
Env_base_1.2.0-07				
Env_base_1.2.0-09				
Env_base_1.2.0-09 (with mca options)				

Details of DIAG routine

(second)

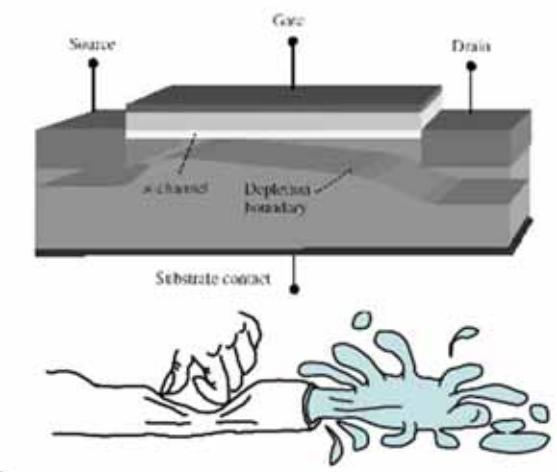
	DIAG	mate	hpsi	pdsyevd	rotv
Env_base_1.2.0-07					
Env_base_1.2.0-09					
Env_base_1.2.0-09 (with mca options)					

We found MPI_BCAST is the downfall

NATORI'S COMPACT MODEL FOR BALLISTIC Si NANOWIRE MOSFET

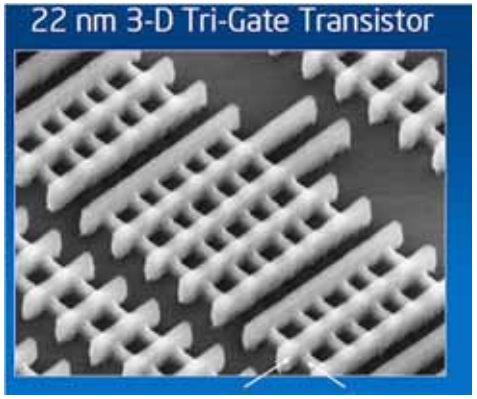
NEW TRANSISTOR STRUCTURES
- SUPPRESSION OF OFF-LEAK CURRENT -

Planar transistor

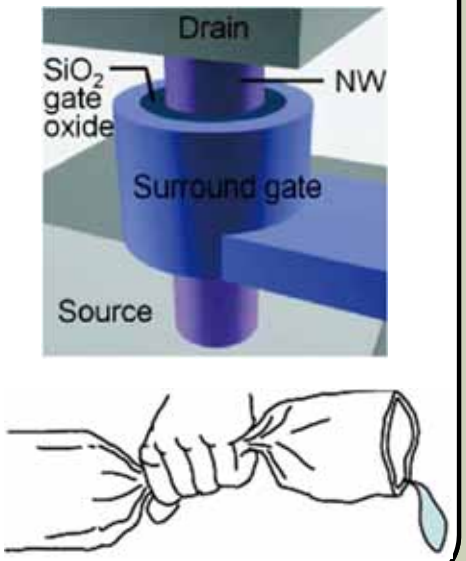


Power consumption by **off-leak current** substantially increases as scaling down of planer FET

tri-gate transistor
Intel Ivy Bridge (2011)



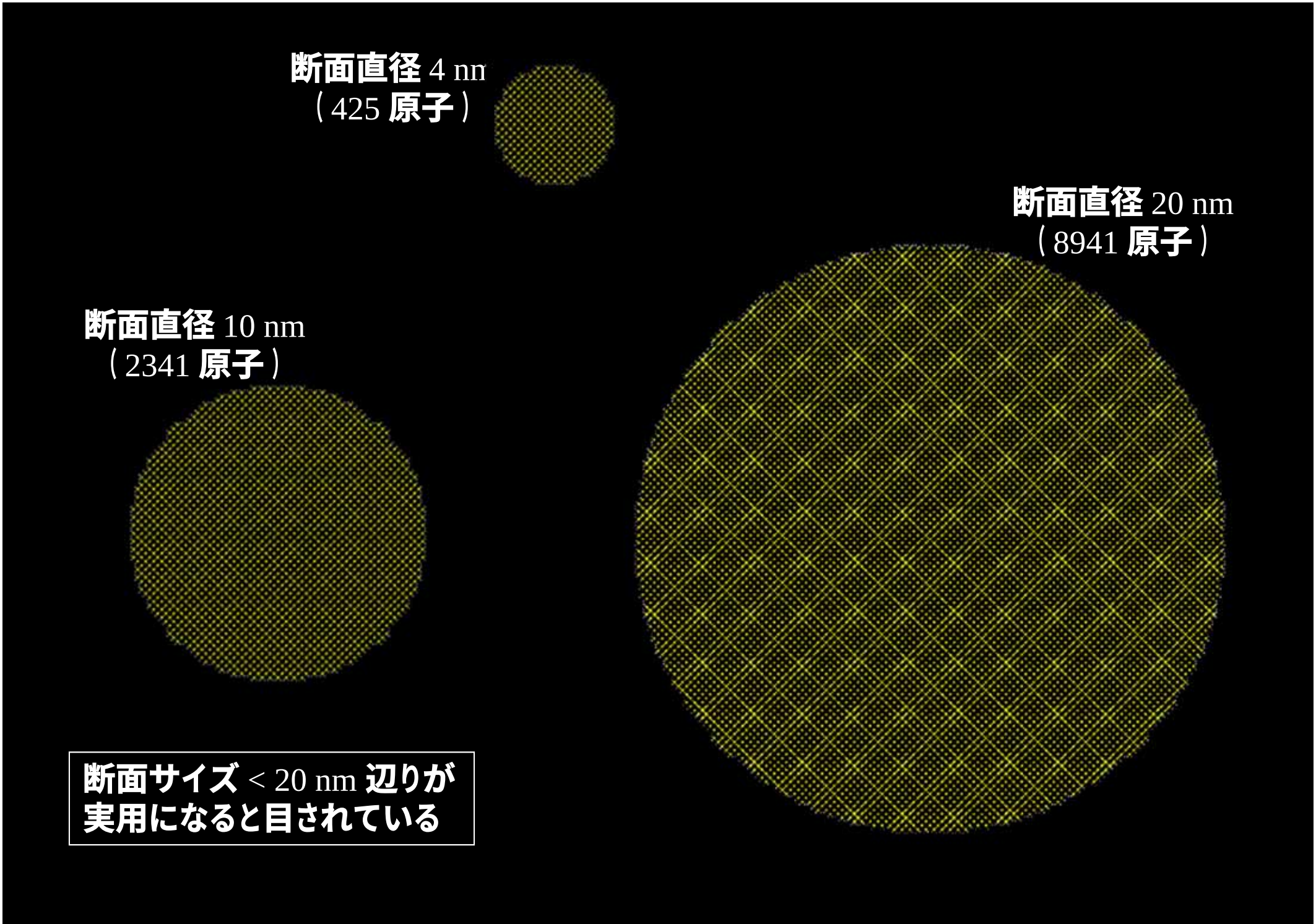
Surrounding-gate transistor



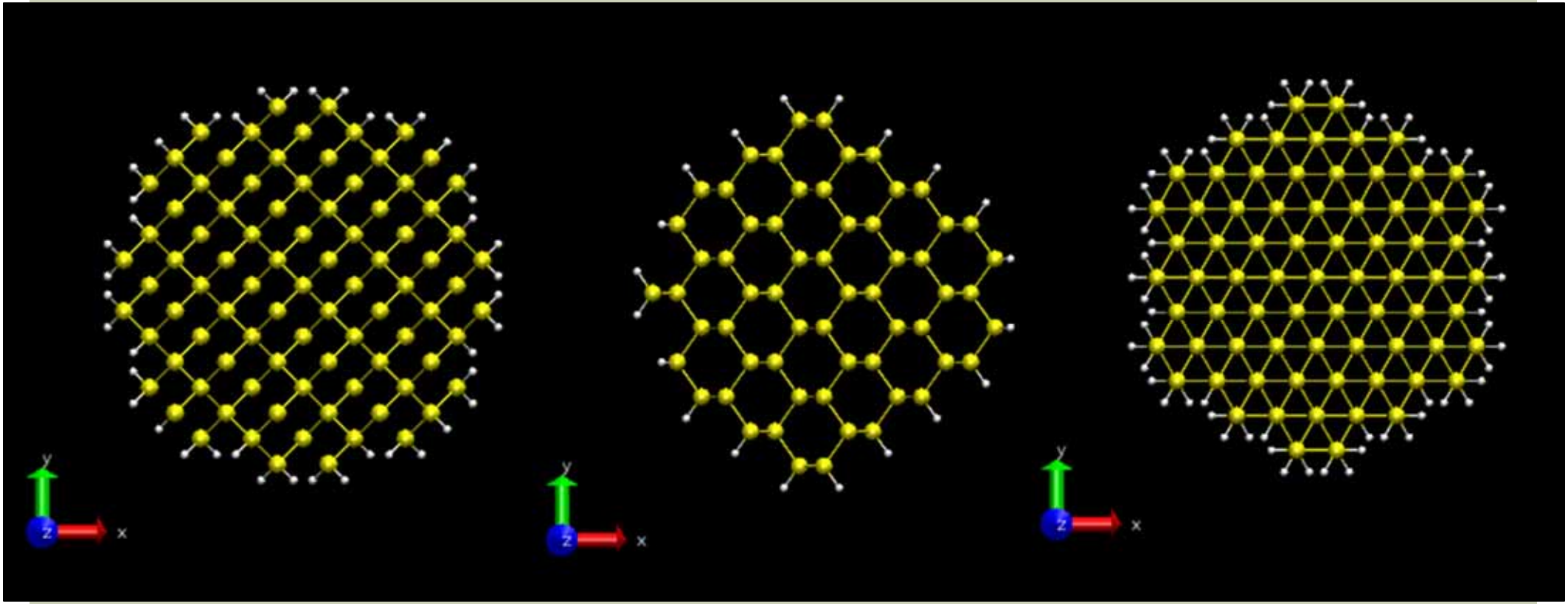
Gate controllability
→ **suppress leaks at off state**
→ **reduce power consumption**

Silicon Nanowire is the most promising channel material for SGFET

シリコンナノワイヤの原子構造　(100)断面



CROSS SECTIONAL VIEWS OF SiNWs



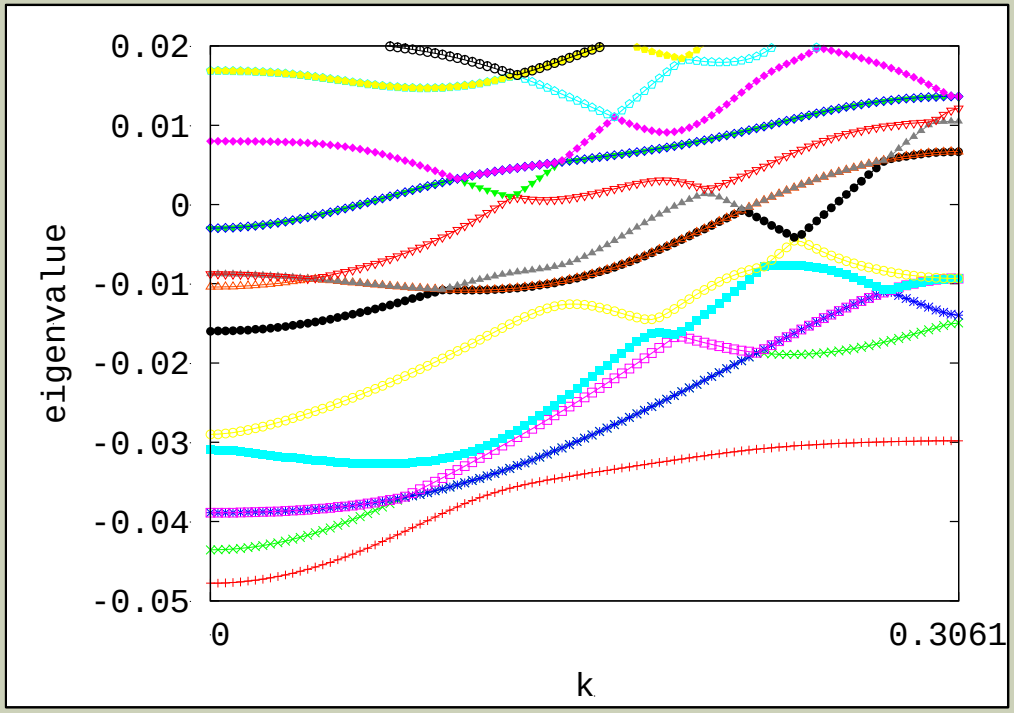
D=1.96nm
[001]

D=1.94nm
[011]

D=1.93nm
[111]

BAND STRUCTURE OF SiNW

$$\left(-\frac{1}{2}\nabla^2 + v_{SCF}(\mathbf{r}) + i\mathbf{k}\cdot\nabla + \frac{k^2}{2}\right)\psi_{n\mathbf{k}}(\mathbf{r}) = \varepsilon_{n\mathbf{k}}\psi_{n\mathbf{k}}(\mathbf{r})$$

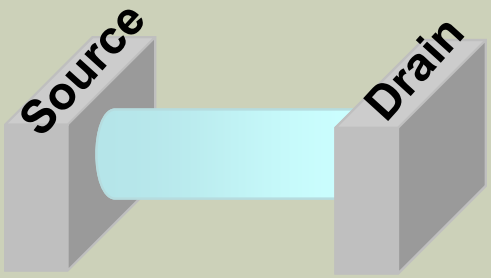


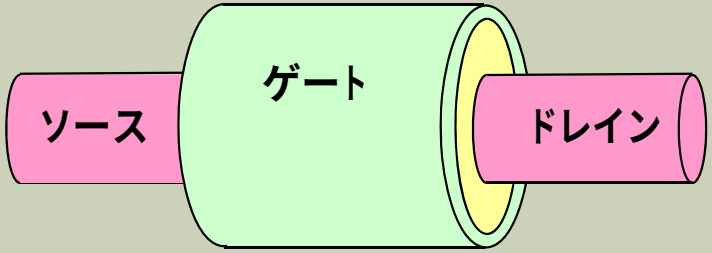
n番目の状態にある電子の速度

$$\frac{d\varepsilon_n(k)}{dk} = \langle \psi_{nk} | -i\nabla | \psi_{nk} \rangle$$

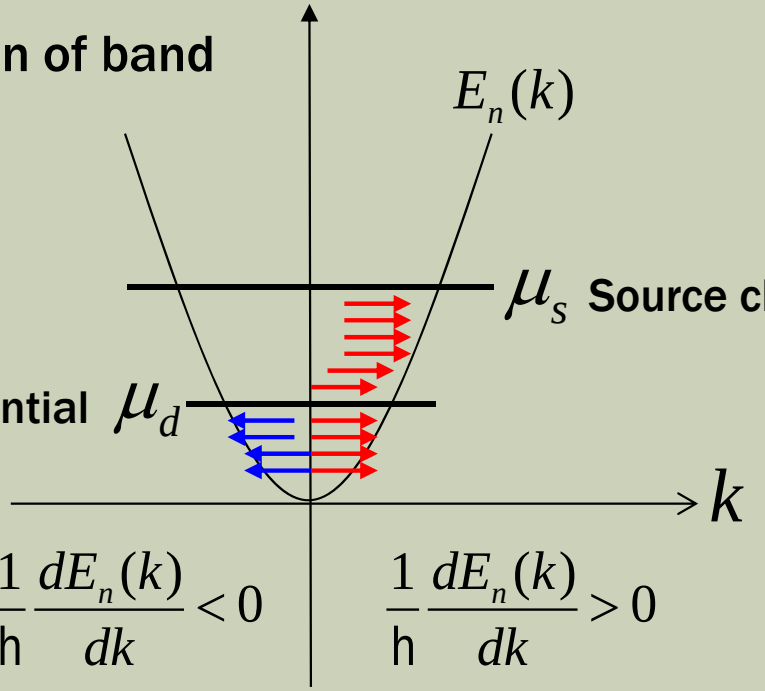
Derivative of the band energy was calculated by r.h.s. of the following formula

コンパクトモデルによる
電流-電圧特性の評価





Schematic illustration of band



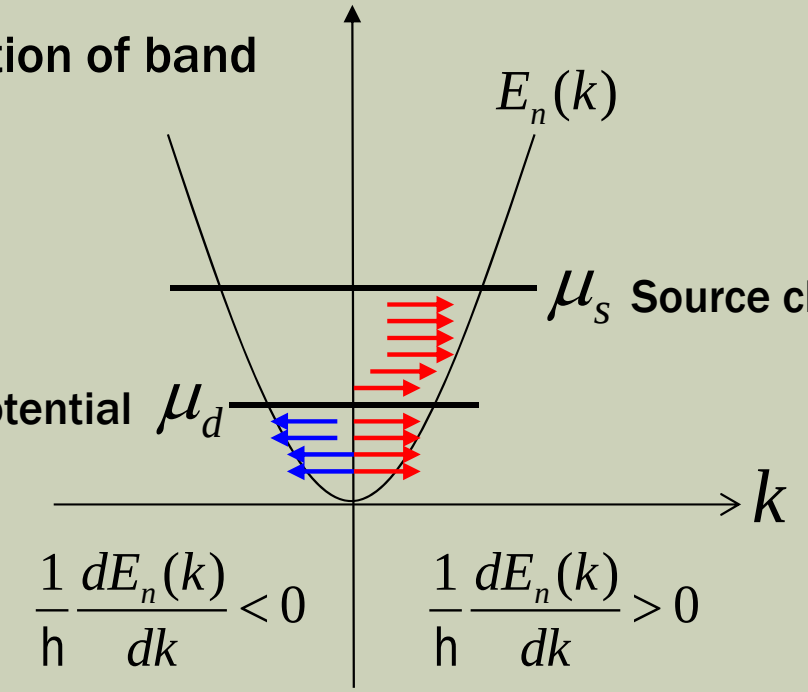
μ_s Source chemical potential

Drain chemical potential μ_d

$$\mu_d = \mu_s - qV_{ds}$$

NUMBER OF CARRIERS IN THE CHANNEL ①

Schematic illustration of band



μ_s Source chemical potential

Drain chemical potential μ_d

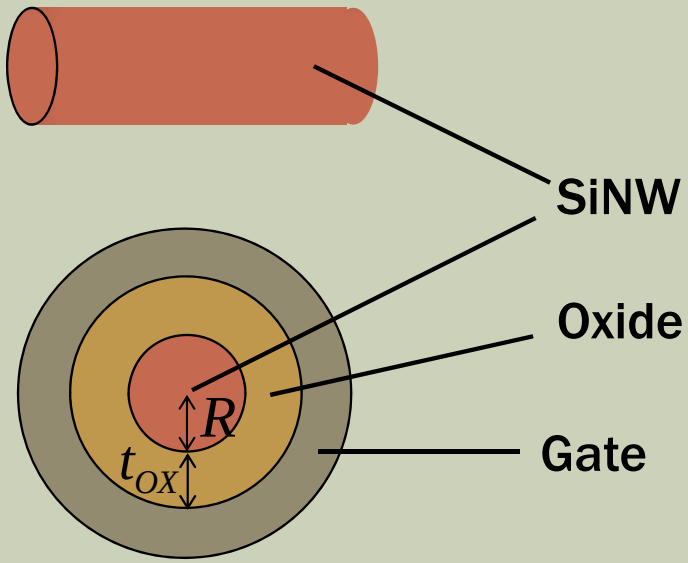
$$\mu_d = \mu_s - qV_{ds}$$

$$N_e^{Fore} = \sum_n 2\int_{\frac{dE_n(k)}{dk}>0} \frac{dk}{2\pi} \frac{1}{1+\exp\left(\frac{E_n(k)-\mu_s}{kT}\right)}$$

$$N_e^{Back} = \sum_n 2\int_{\frac{dE_n(k)}{dk}<0} \frac{dk}{2\pi} \frac{1}{1+\exp\left(\frac{E_n(k)-\mu_d}{kT}\right)}$$

$$N_e^{Channel} = N_e^{Fore} + N_e^{Back}$$

NUMBER OF CARRIERS IN THE CHANNEL ②



$$Q_e^{Channel} = q\left(N_e^{Fore} + N_e^{Back}\right)$$

$$C_g = \frac{\varepsilon_{OX}}{2\ln\left(\frac{R+t_{OX}}{R}\right)}$$

$$Q_e^{Channel} = C_g \phi_g$$

$$\phi_g = V_g - V_{th} - \frac{\mu_s - E_{CBM}}{q}$$

CURRENT FORMULA

$$I_d = 2q\sum_n \int_{\frac{dE_n(k)}{dk}>0} \frac{dk}{2\pi}\frac{1}{\mathrm{h}}\frac{dE_n(k)}{dk}\frac{1}{1+\exp\left(\frac{E_n(k)-\mu_s}{kT}\right)}$$

$$+2q\sum_n \int_{\frac{dE_n(k)}{dk}<0} \frac{dk}{2\pi}\frac{1}{\mathrm{h}}\frac{dE_n(k)}{dk}\frac{1}{1+\exp\left(\frac{E_n(k)-\mu_d}{kT}\right)}$$

$$= \frac{q}{\pi\mathrm{h}}\sum_n \int dE_n \frac{1}{1+\exp\left(\frac{E_n-\mu_s}{kT}\right)} - \frac{q}{\pi\mathrm{h}}\sum_n \int dE_n \frac{1}{1+\exp\left(\frac{E_n-\mu_d}{kT}\right)}$$

$$= \frac{q}{\pi\mathrm{h}}\sum_n \int dE_n \left(f(E_n,\mu_s)-f(E_n,\mu_d)\right)$$

Landauer formula

$$I = \frac{q}{\pi\mathrm{h}}\sum_n T_n(E)\int dE\left(f(E,\mu_s)-f(E,\mu_d)\right)$$

CALCULATION PROCEDURE ①

① **Perform band calculation**

$$\left(-\frac{\hbar^2}{2m}\nabla^2 + v(\mathbf{r})\right)\psi_{nk}(\mathbf{r}) = E_n(k)\psi_{nk}(\mathbf{r})$$

Get

$$\begin{cases} E_n(k) \\ \dfrac{1}{\hbar}\dfrac{dE_n(k)}{dk} = \dfrac{1}{m}\left\langle \psi_{nk}\middle| -i\hbar\nabla \middle|\psi_{nk}\right\rangle \end{cases}$$

② **Give V_g-V_{th} and V_{ds} as input parameters, and solve the following equation**

$$\begin{cases} C_g\left(V_g - V_{th} - \dfrac{\mu_s - E_{CBM}}{q}\right) = Q_e^{Channel} = q\left(N_e^{Fore} + N_e^{Back}\right) \\ N_e^{Fore} = \displaystyle\sum_n 2\int_{\frac{dE_n(k)}{dk}>0} \frac{dk}{2\pi}\frac{1}{1+\exp\left(\dfrac{E_n(k)-\mu_s}{kT}\right)} \\ N_e^{Back} = \displaystyle\sum_n 2\int_{\frac{dE_n(k)}{dk}<0} \frac{dk}{2\pi}\frac{1}{1+\exp\left(\dfrac{E_n(k)-\mu_d}{kT}\right)} \\ \mu_d = \mu_s - qV_{ds} \end{cases}$$

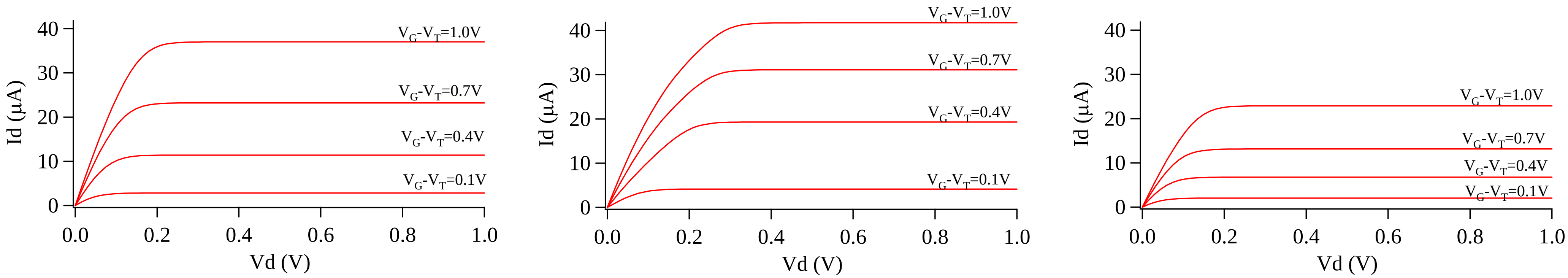
→ **Get** μ_s

CALCULATION PROCEDURE ②

③ Calculate the drain current by the following formula

$$I_d = 2q\sum_n \int_{\frac{dE_n(k)}{dk}>0} \frac{dk}{2\pi}\frac{1}{\mathrm{h}}\frac{dE_n(k)}{dk}\frac{1}{1+\exp\left(\frac{E_n(k)-\mu_s}{kT}\right)}$$

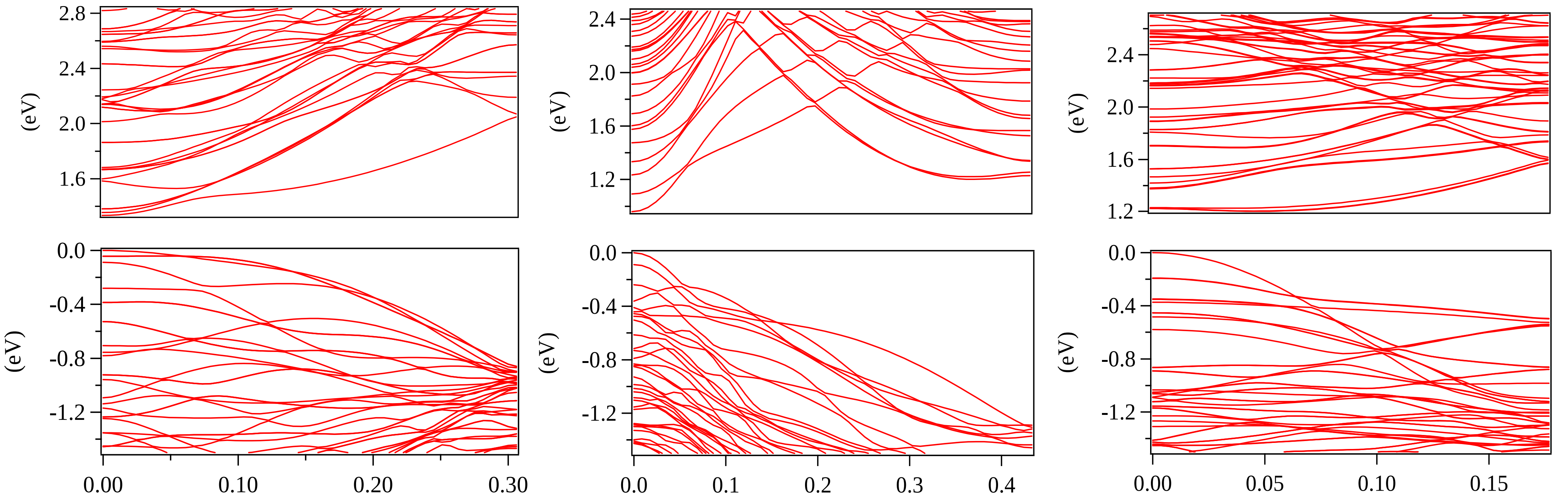
$$+2q\sum_n \int_{\frac{dE_n(k)}{dk}<0} \frac{dk}{2\pi}\frac{1}{\mathrm{h}}\frac{dE_n(k)}{dk}\frac{1}{1+\exp\left(\frac{E_n(k)-\mu_d}{kT}\right)}$$

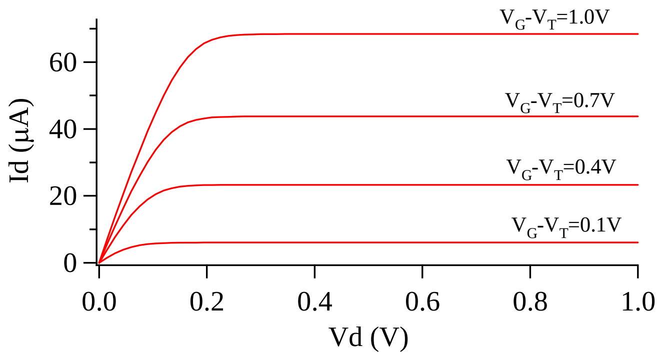


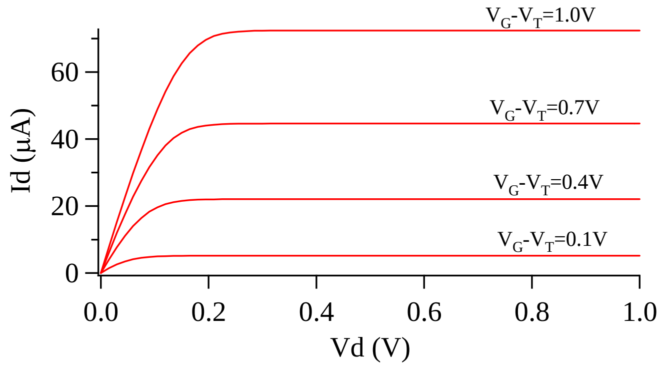
SiNW(100)
diameter：2nm
temperature：300K

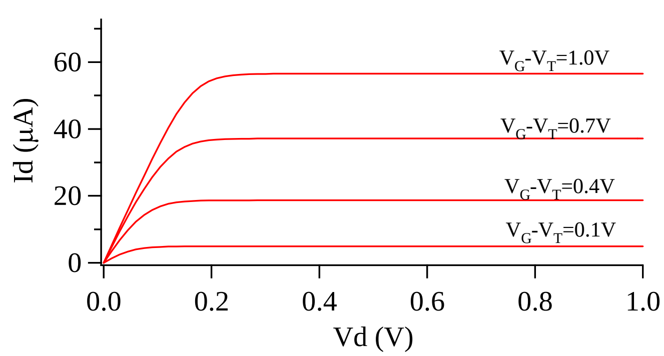
SiNW(110)
diameter：2nm
temperature：300K

SiNW(111)
diameter：2nm
temperature：300K





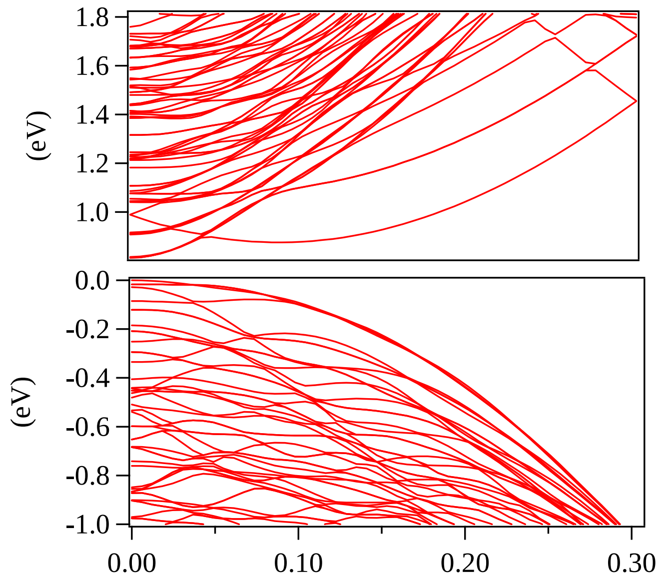


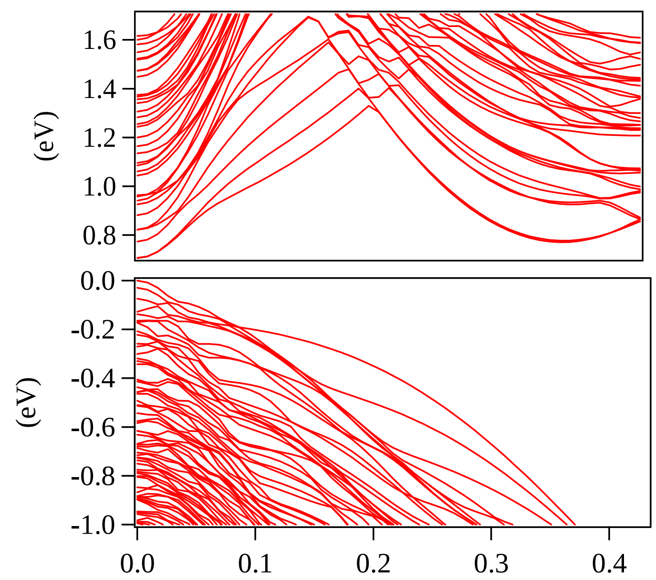


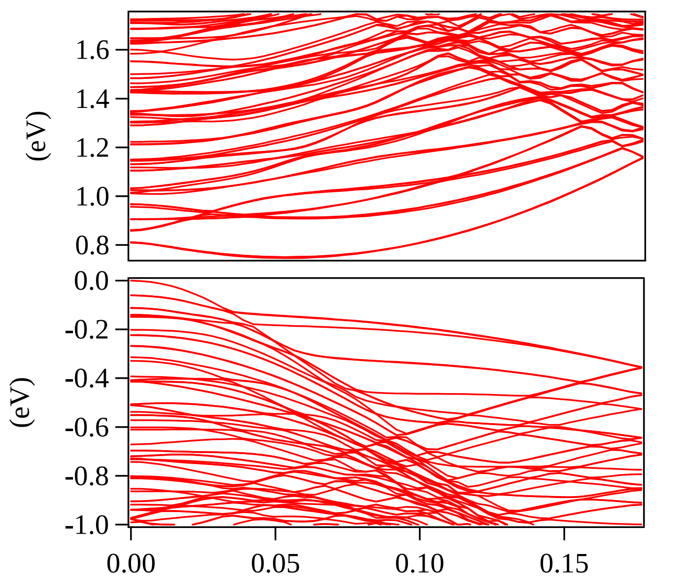
SiNW(100)
diameter：4nm
temperature：300K

SiNW(110)
diameter：4nm
temperature：300K

SiNW(111)
diameter：4nm
temperature：300K

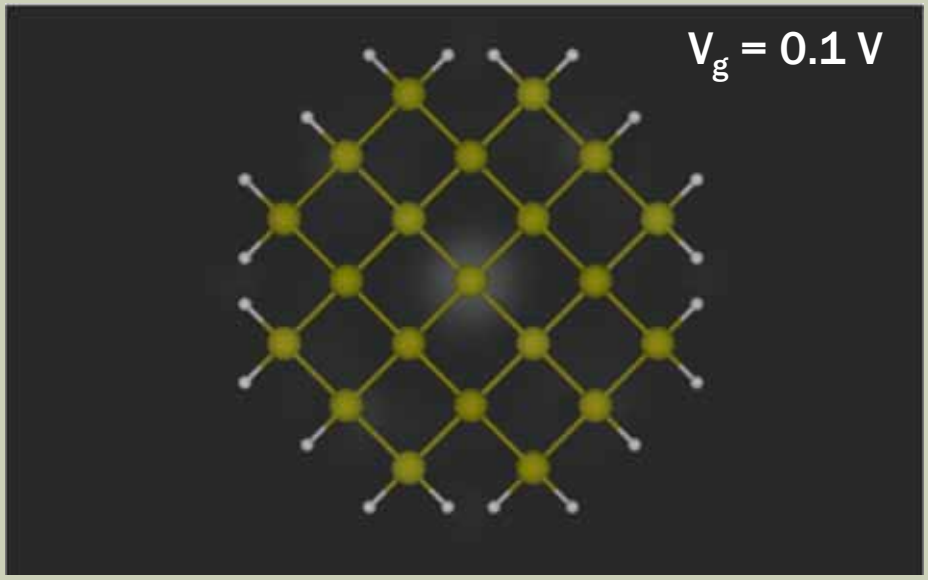


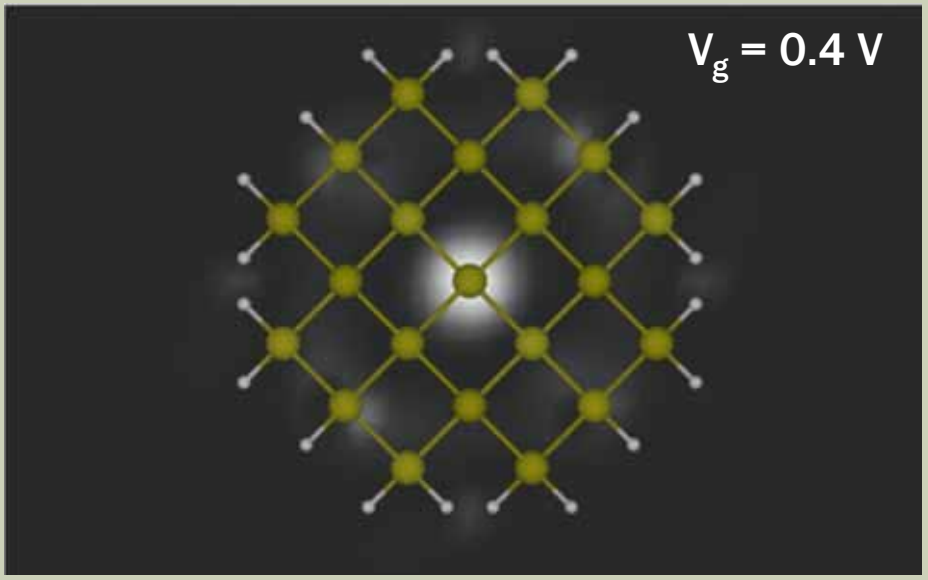


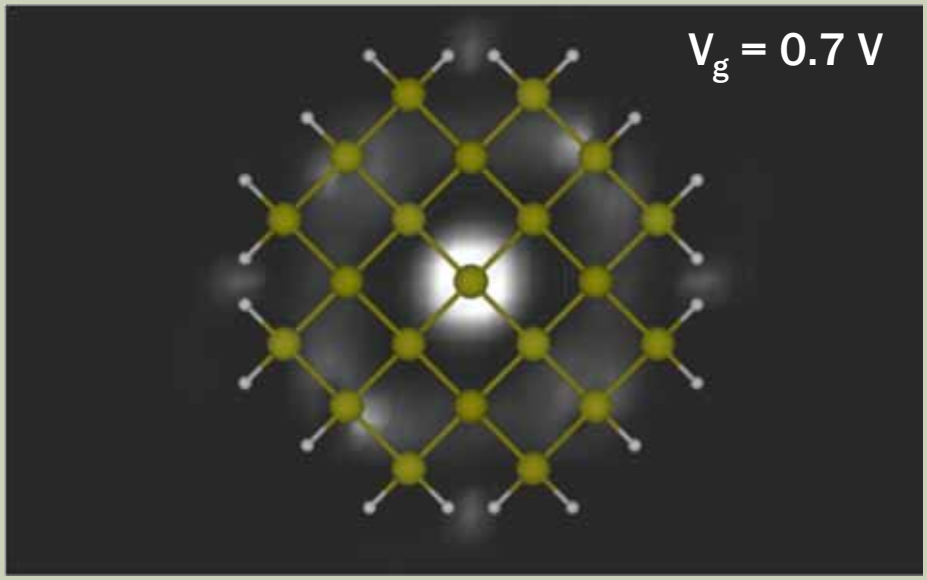


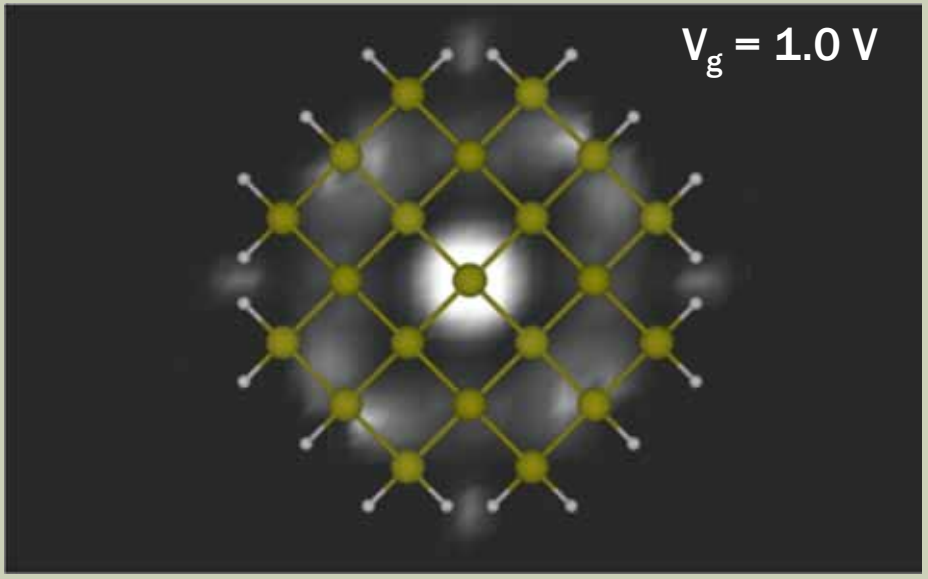
CONTOUR PLOT OF CURRENT DENSITY

By S. Furuya (VESTA)





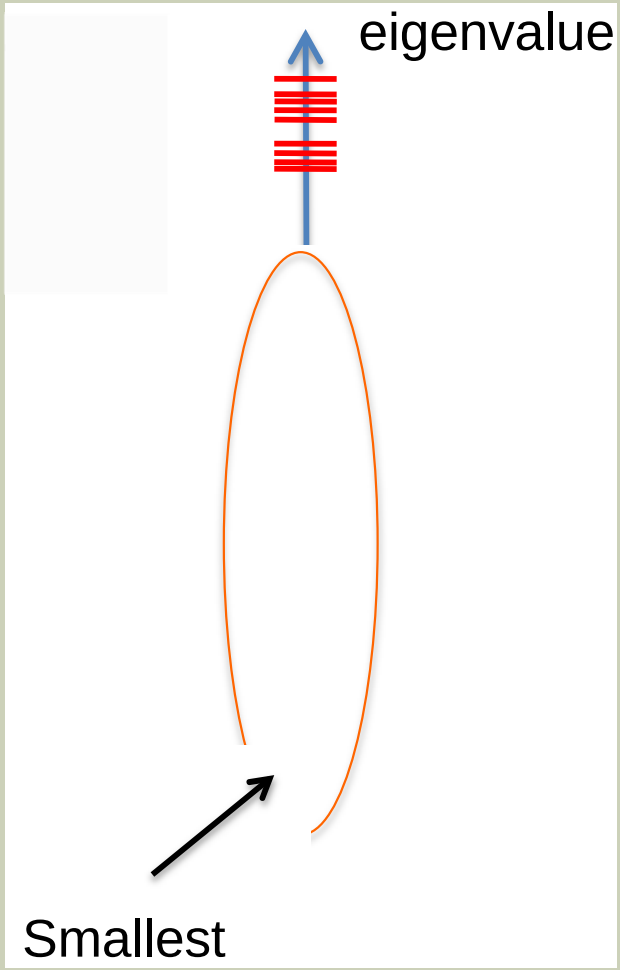




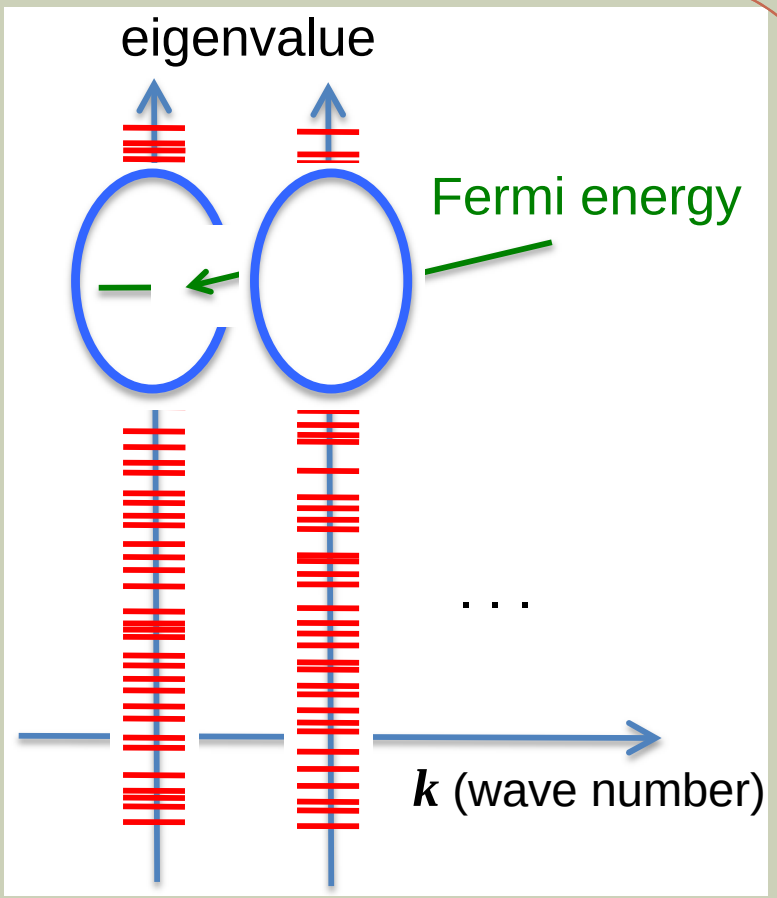
BAND STRUCTURE CALCULATIONS WITH SAKURAI-SUGIURA METHOD

二村保徳
筑波大
櫻井研

SCF CALCULATIONS AND BAND CALCULAITONS



SCF calc.→ exterior eigenproblem



Band calc.→interior eigenproblem

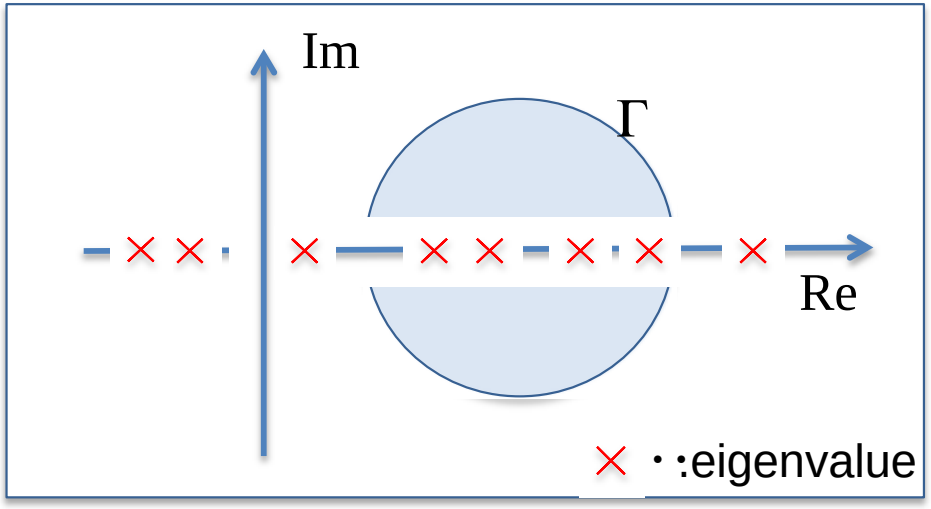
SAKURAI-SUGIURA METHOD

- A novel eigensolver
  - Suitable for interior eigenproblems
  - Suitable for massively-parallel architectures
- Band structure calculation of 10,000-atom system

CONTOUR INTEGRATION

$$S_k \equiv \frac{1}{2\pi \mathrm{i}} \int_\Gamma z^k \left(zI - A\right)^{-1} V \, \mathrm{d}z$$

L arbitrary vectors
(linearly independent)



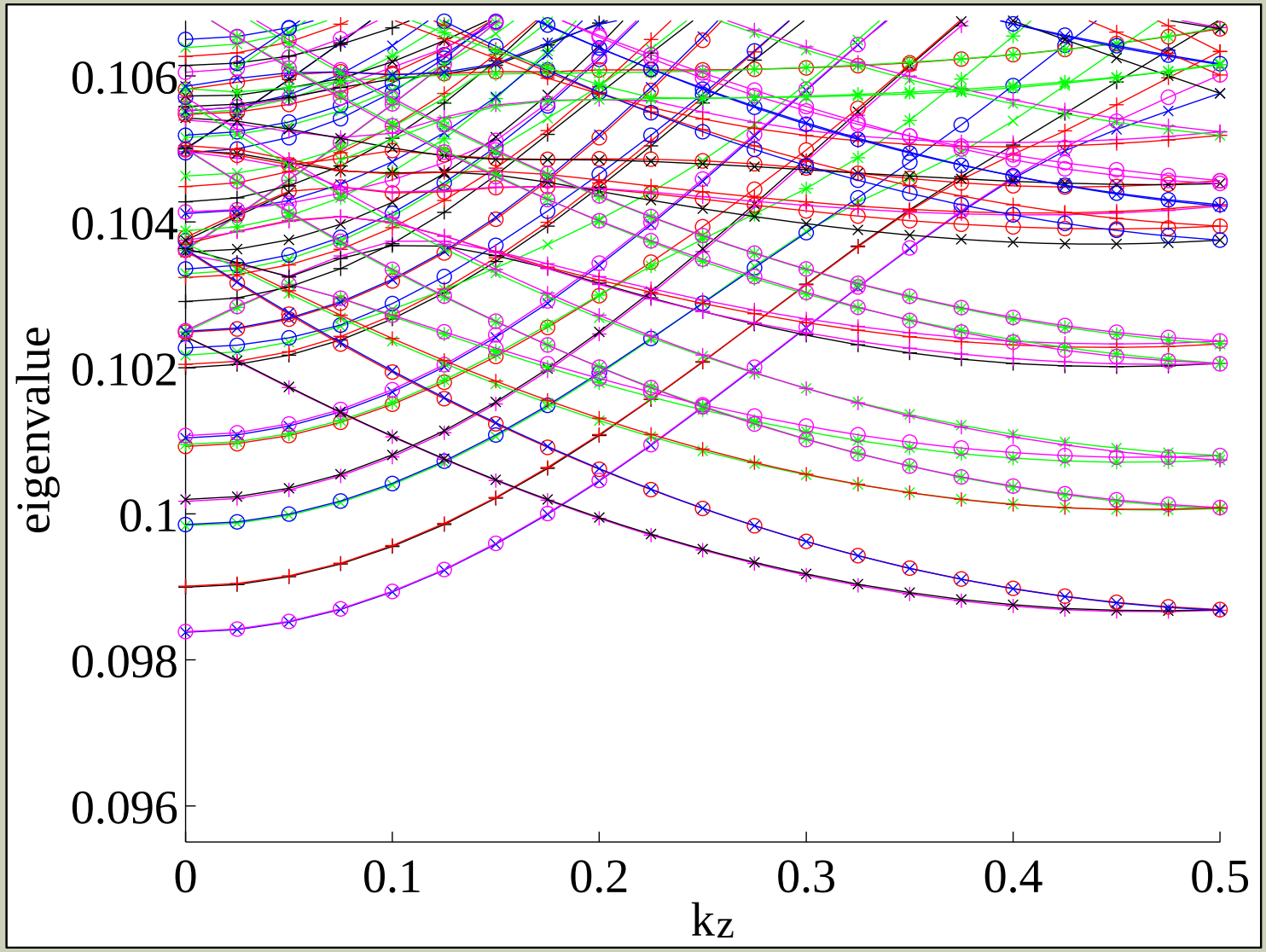
$S_k, V \in \mathbb{C}^{n \times L}$

$k = 0, 1, \ldots, M-1$

$L, M << n$

# of grid points

BAND STRUCTURE OF 10,000-ATOM SiNW

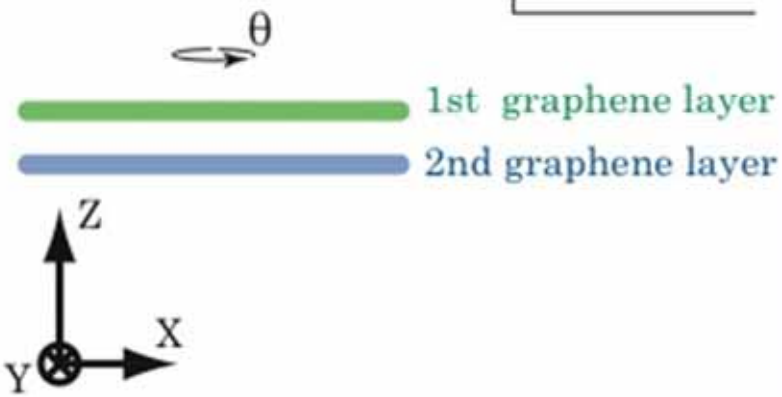


ATOMIC & ELECTRONIC STRUCTURES OF TWISTED BILAYER GRAPHENE

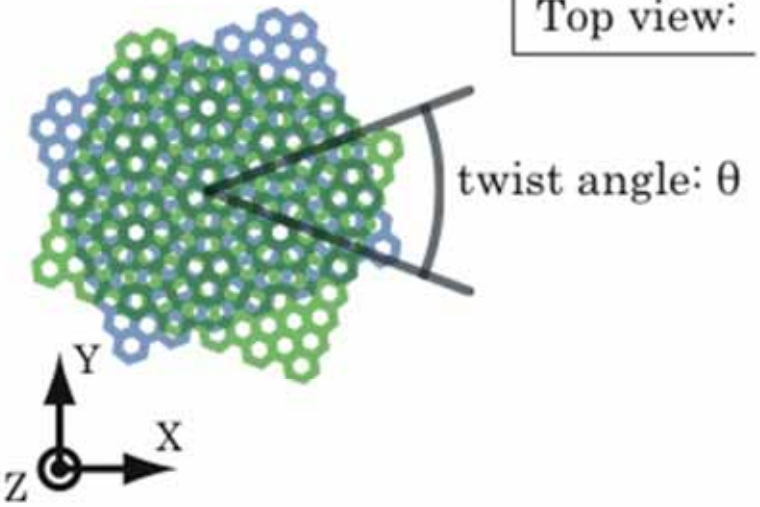
内田和之
東大押山研

What is Twisted Bilayer Graphene (tBLG)?

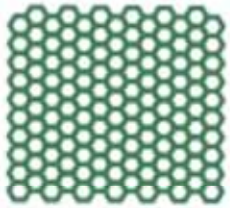
Side view:



Top view:



■ θ=0° : AA-stacking



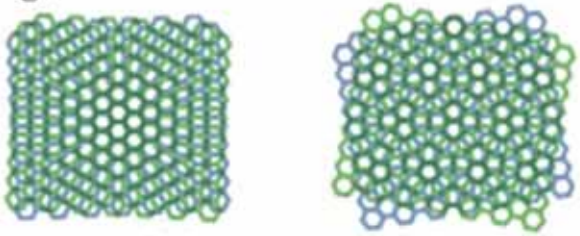
4 atom/cell

■ θ=60° : AB-stacking

4 atom/cell

■ θ=any value(≠0°,60°)

(e.g.) θ=6° θ=20°



"Moire pattern"

- periodic only for special θs (→this work)
- non-periodic for other θs

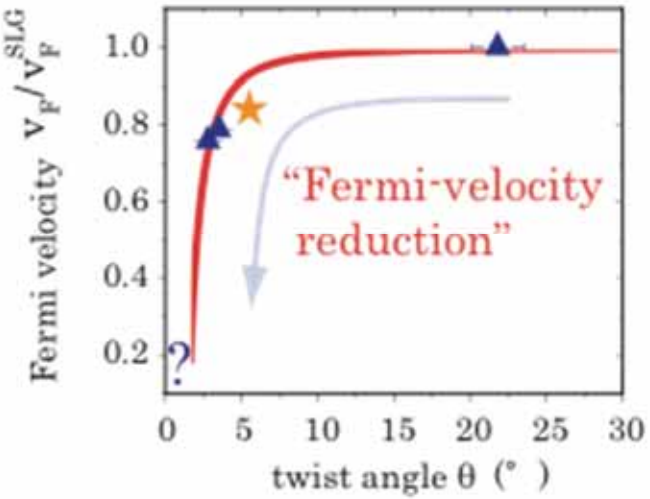
*Assumption: In the above and following, an A-site atom is on the twist axis in each graphene. Other cases will be also discussed later (consideration of lateral translations).

Why tBLG?

■ ①STM+STS for Twisted Graphene Layers

Luican et al. : PRL 106, 126802 (2011)

(STM→ Moire → twist angle : θ
STS under magnetic field→Landau levels→Fermi velocity : V_F)



▲ : STM+STS
— : continuum tight-binding calc.
★ : DFT calc
[Trambly et al., Nano Lett 10, 804 (2010)]

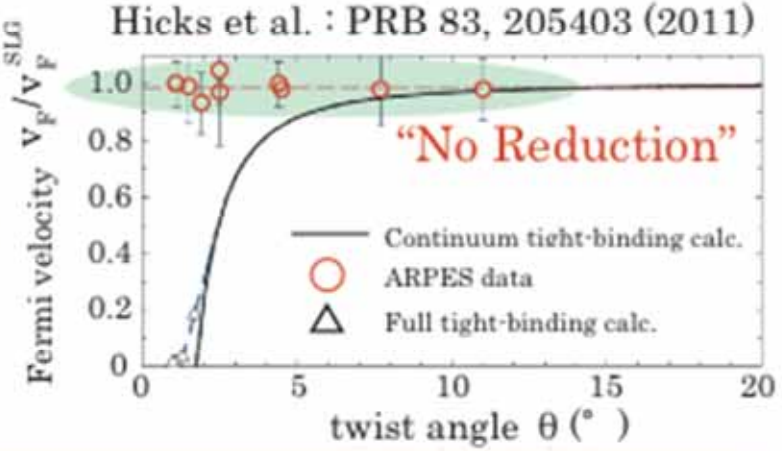
■ Fermi velocity V_F is controlled by changing twist angle θ.

■ consistent with tight-binding calc. for tBLGs

However, controversial

■ ②ARPES

Hicks et al. : PRB 83, 205403 (2011)

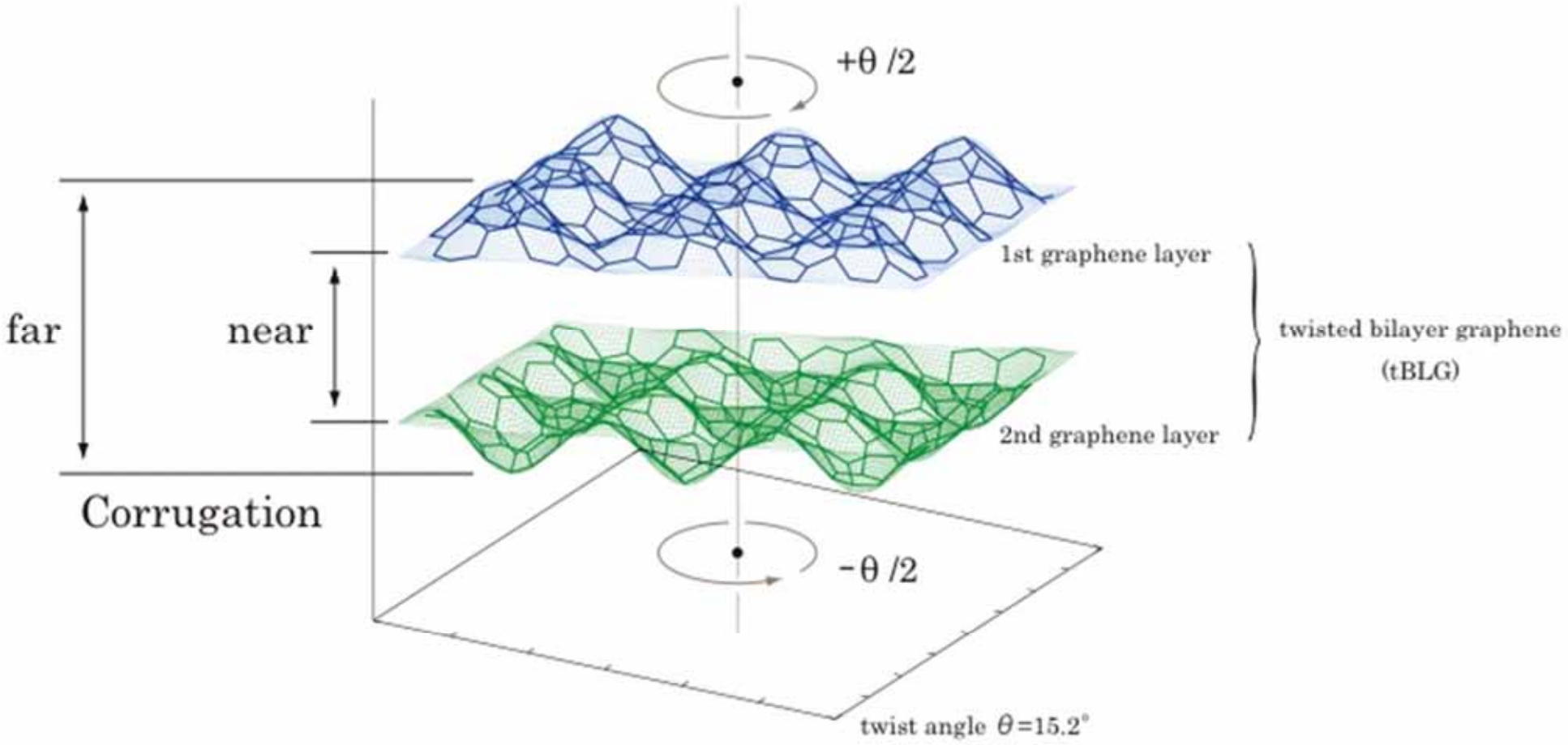


THIS WORK :

We wish to know if the Fermi-velocity reduction is true or not.

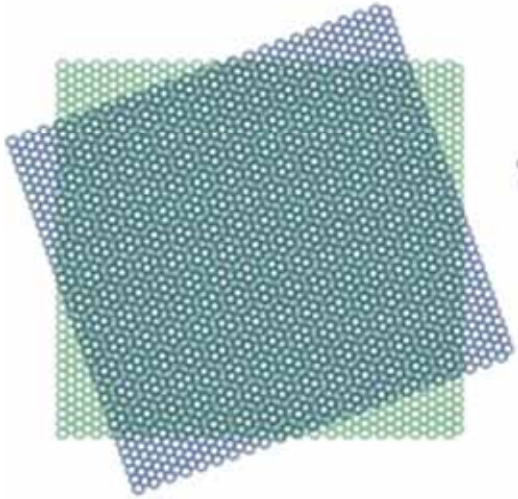
→ DFT calculations for tBLGs

Atomic-structure Optimization

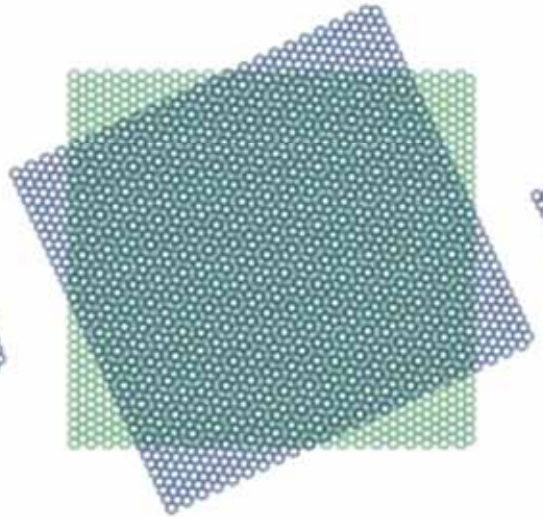


Relation between Corrugations and Stacking Structures

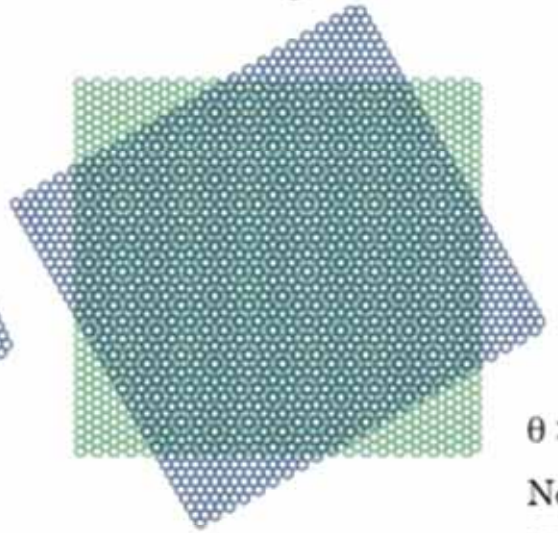
■twist angle θ=30°



■twist angle θ=25°



■twist angle θ=20°

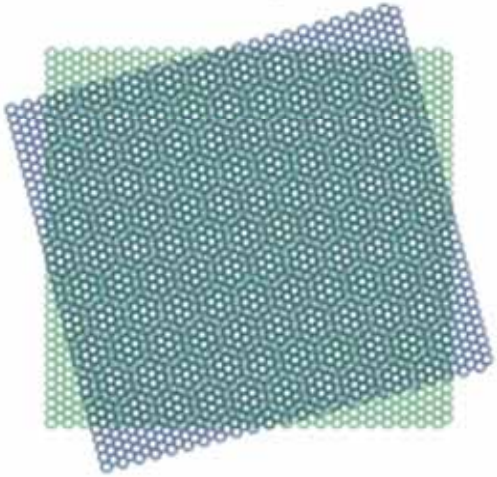


θ > ~20°

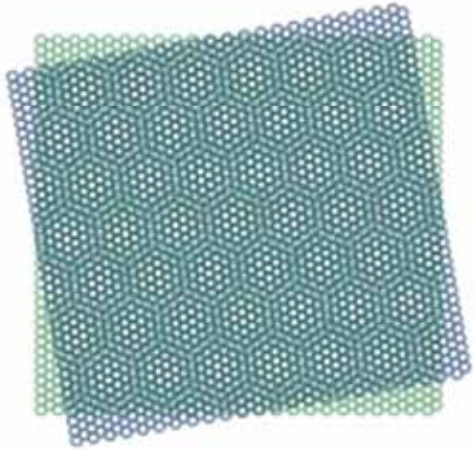
No Corrugation

No locally AA/AB stacked regions

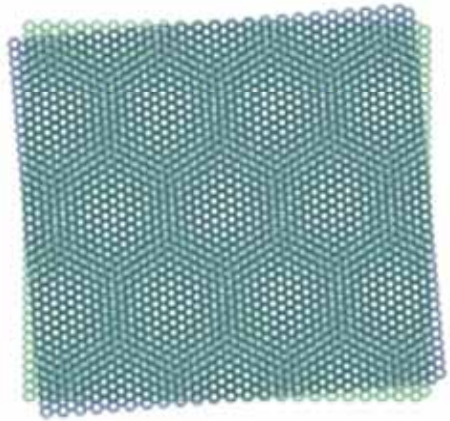
■twist angle θ=15°



■twist angle θ=10°



■twist angle θ=5°



θ < ~20°

Corrugation

Locally AA/AB stacked regions exist, and become larger as θ becomes smaller

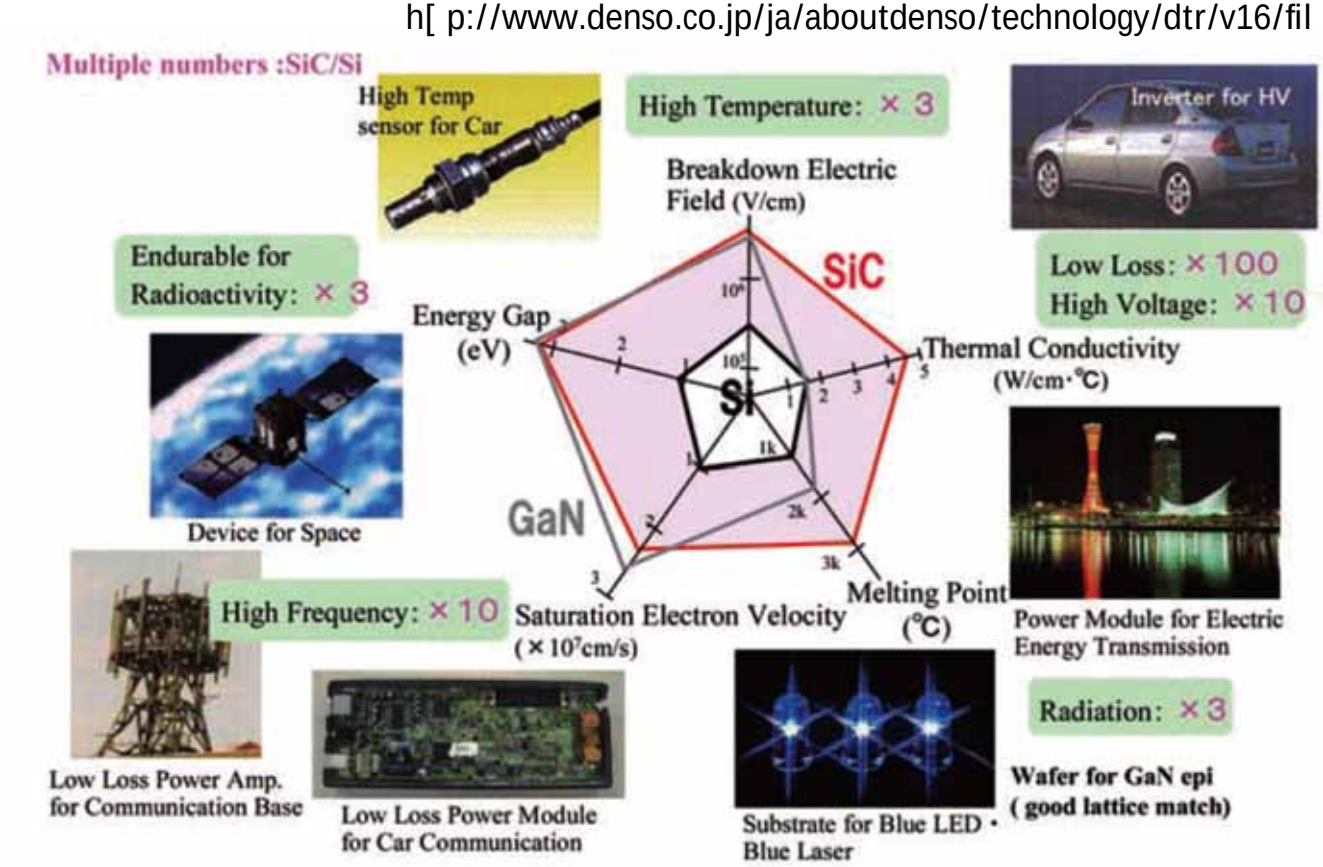
FIRST-PRINCIPLES CALCULATIONS OF STEP STRUCTURES ON SiC(0001)

澤田啓介
東大押山研

Silicon-Carbide

- Silicon carbide (SiC) is a hopeful semiconductor for the next genera4on of **power-electronic-device**.

h[p://www.denso.co.jp/ja/aboutdenso/technology/dtr/v16/fil s/13.pdf

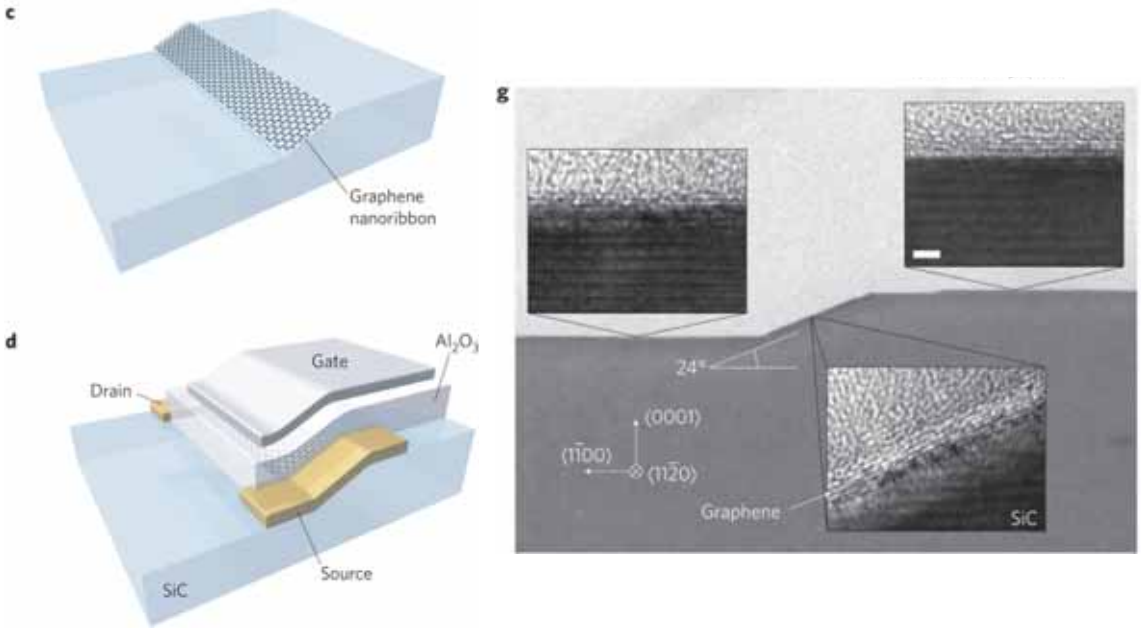


Features and applica4ons of SiC semiconductors.

Template-of-Nanostructures-using-Step-Structures

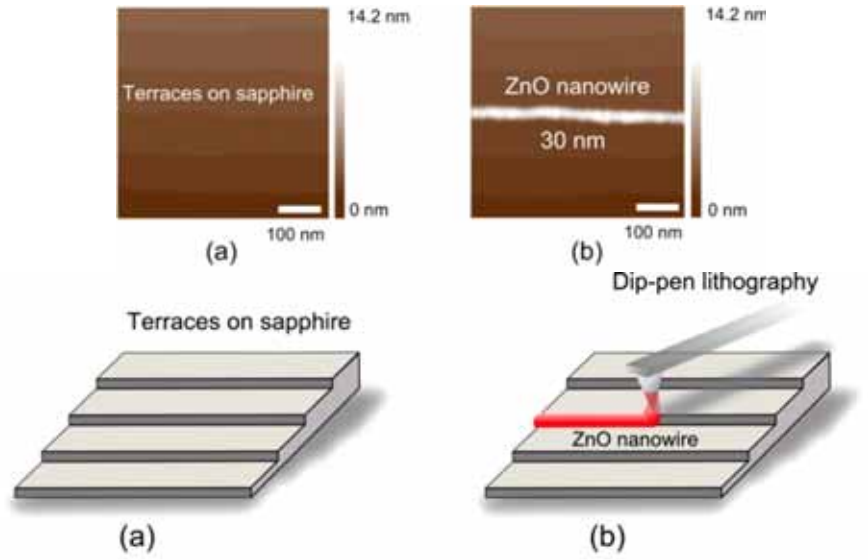
- Step structures can be applicable to the **template-of-nanostructures** such as nano wires, tubes and dots.

M. Sprinkle et al., Nat. Nanotechnol. **5**, 727 (2010).



Nanofacet on 4H-SiC(0001) is applied to template of the field-effect transistor using self-organized graphene nanoribbon.

J. Y. Son et al., Electrochem. Solid-State Lett. **14**, H397 (2011).



The fabrication of ZnO nanowires is demonstrated on steps of sapphire [Al_2O_3 (0001)] surface.

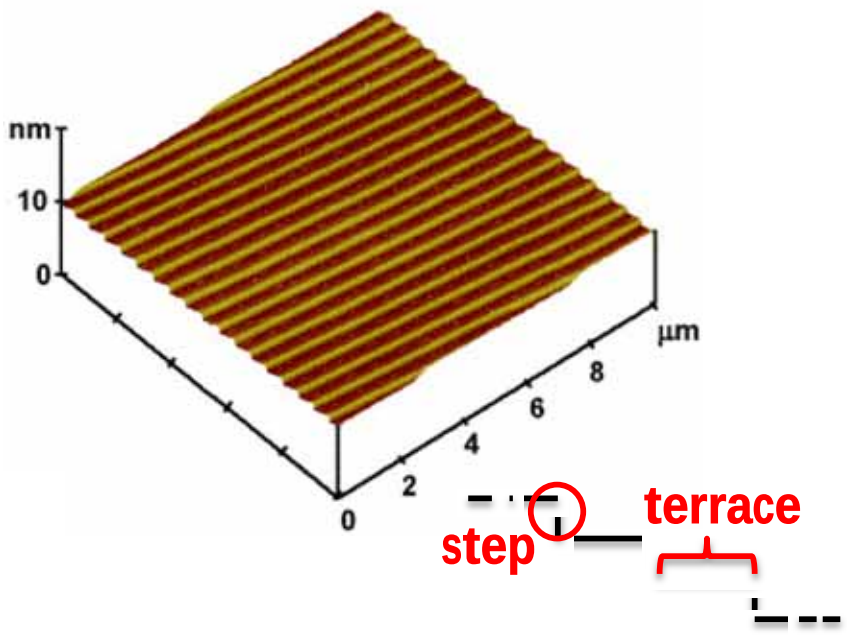
- SiC基板の熱分解によるグラフェン生成

Morphologies-of-Vicinal-Solid-Surface

- It is important to understand structures of surfaces or interfaces on substrates for the device fabrications.
- Vicinal solid surface often shows regularly spaced **step,-terrace-and-nanofacet-**formed by step bunching.

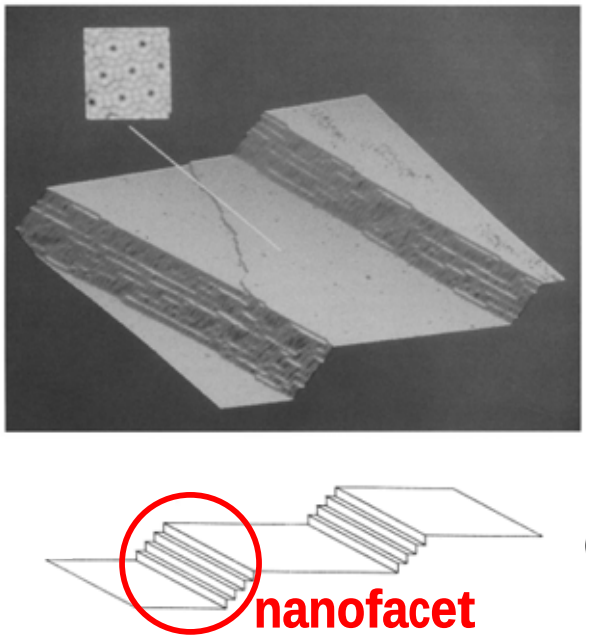
A. Nakajima et al., J. Crystal Growth **278**, 437 (2005).

AFM image of 6H-SiC substrate surface.



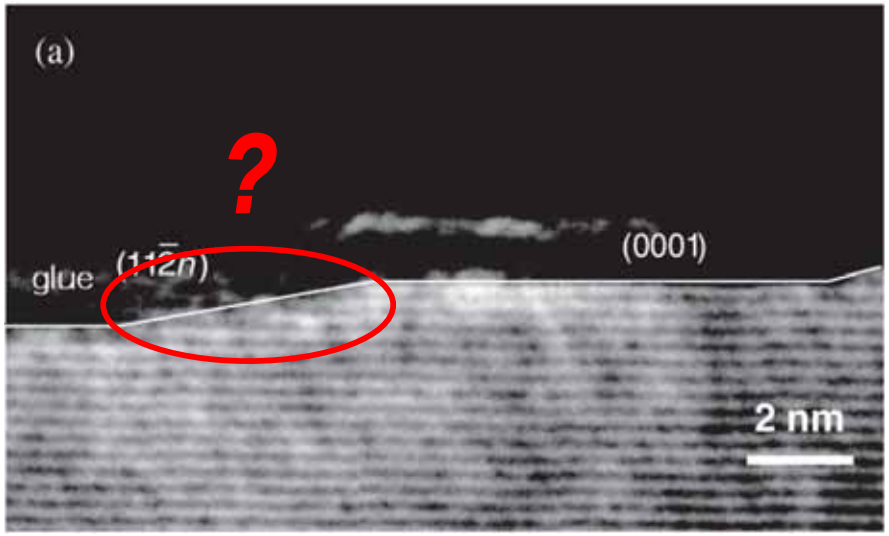
E. D. Williams et al., Surf. Sci. **294**, 219 (1993).

STM image of Si(111) surface.

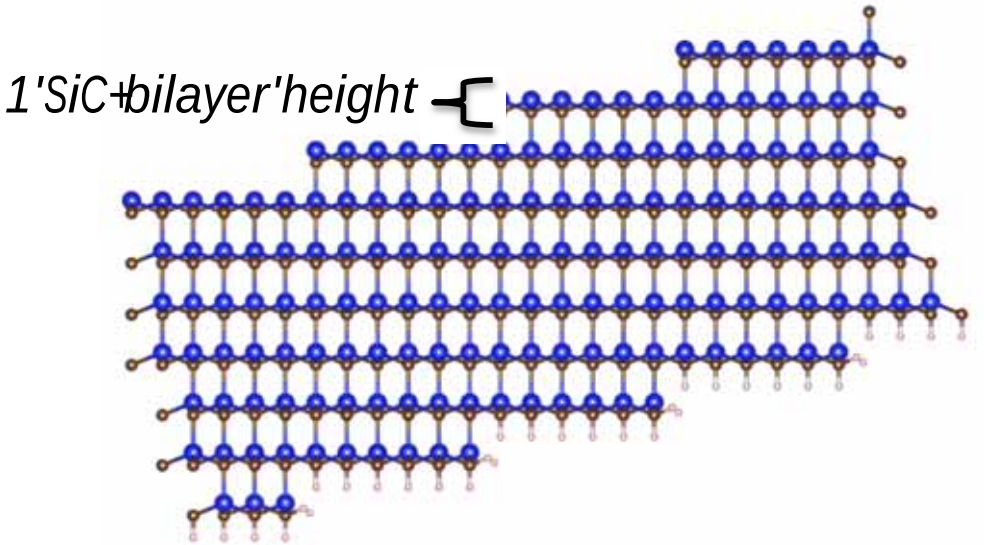


Expected-Simple-Step-Structures

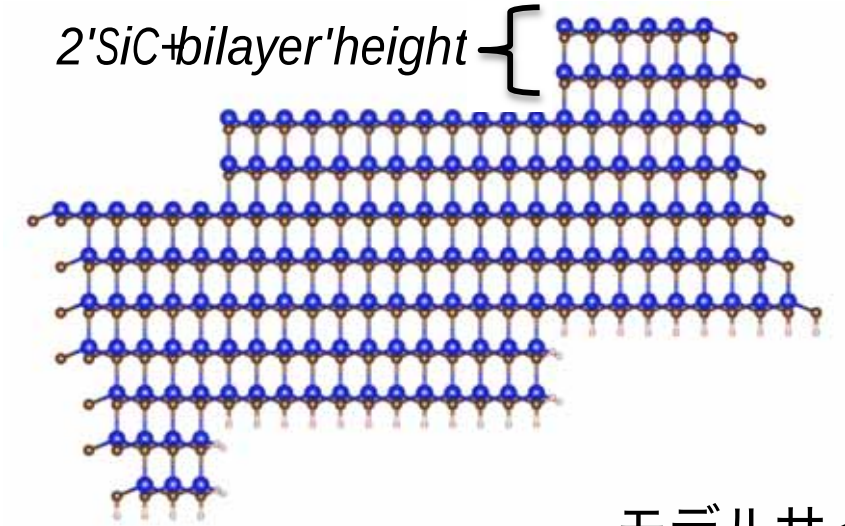
Cross-sectional-TEM-image



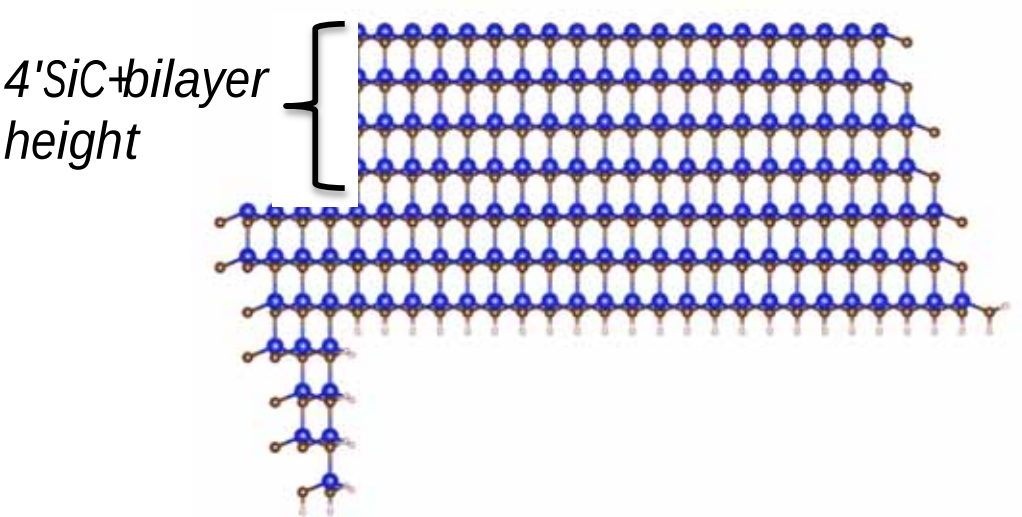
Single-height-step-(SHS)-structure-



Double-height-step-(DHS)-structure-



Quad-height-step-(QHS)-structure



モデルサイズ：300～1000原子

まとめと今後

- コンパクトモデルを用いたSiNW-FETの電流電圧特性の計算
  - ゲート電極の効果を第一原理的に扱う（産総研 大谷実）
  - ソース・ドレイン電極も取り入れた第一原理輸送計算
    （東大押山研 Zixin Guo、東大渡邊研 笹岡健二）
  - 第一原理デバイスシミュレータ
- 捻れ二層グラフェンの構造と電子状態（東大押山研 内田和之）
- SiC微斜面の構造決定（東大押山研 澤田啓介）
- 機能拡張
  - RSDFT-CPMDの実装（東大押山研 小泉健一、阪大 重田照育）