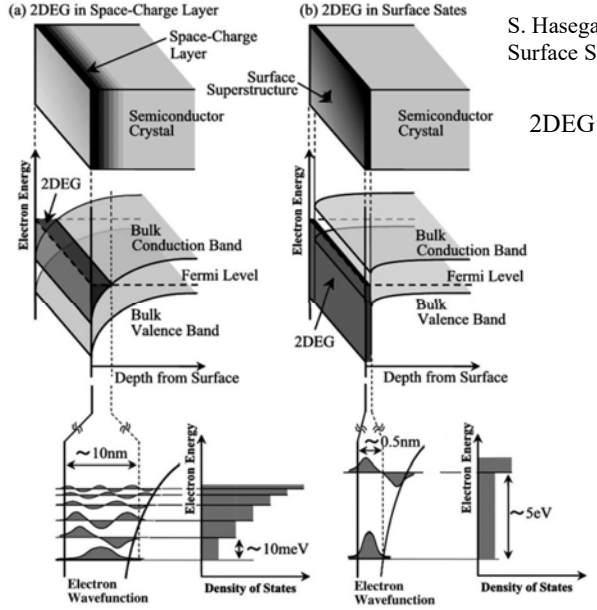
Schedule of The First Half of The Course (Hasegawa)

Lecture Slides (PDF files)
http://www-surface.phys.s.u-tokyo.ac.jp/KougiOHP/

1. Nanoscience and Surface Physics ナノサイエンスと表面物理
   Nanoscience in Nobel Prize
2. Atomic Arrangements at Surfaces 表面原子配列構造
   Scanning Tunneling Microscopy, Electron Diffraction
   走査トンネル顕微鏡、電子回折
3. Surface Electronic States 表面電子状態
   Surface states 表面状態、 Rashba Effect ラシュバ効果
   Topological Surface States トポロジカル表面状態、
   Band Bending バンド湾曲
4. Surface Electronic Transport 表面電気伝導
   Space-Charge-Layer Transport and Surface-State Transport
   空間電荷層伝導と表面状態伝導
   2D Materials 2次元物質
   Atomic-Layer Superconductivity 原子層超伝導

Surface States and Space-Charge Layer (Band bending)

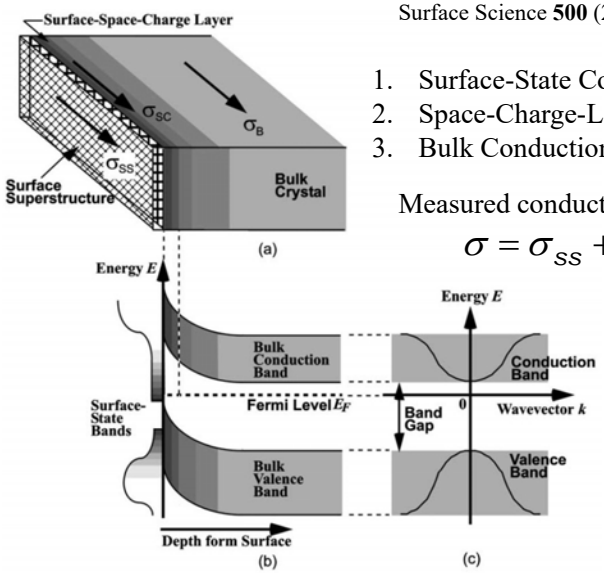


S. Hasegawa & F. Grey,
Surface Science **500** (2002) 84–104

2DEG: Two-dimensional Electron Gas

Three Channels for Electrical Conduction

S. Hasegawa & F. Grey,
Surface Science **500** (2002) 84–104



1. Surface-State Conduction
2. Space-Charge-Layer Conduction
3. Bulk Conduction

Measured conductivity

$$\sigma = \sigma_{ss} + \sigma_{sc} + \sigma_B$$

電界効果トランジスタ Field-Effect Transistor FET

Gate-Controlling
Bending of Bulk Bands near Surface

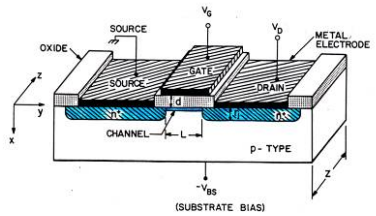
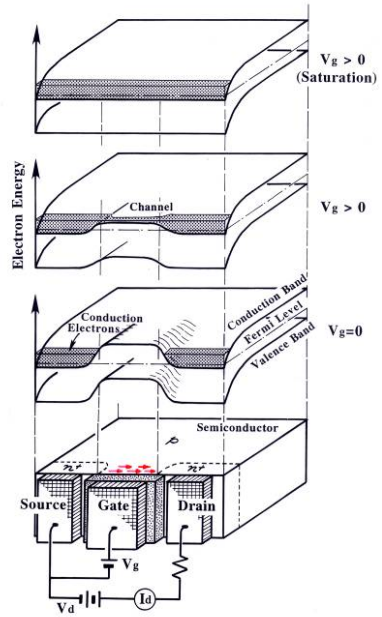


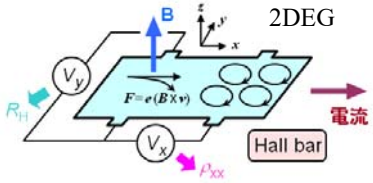
Fig. 3 Schematic diagram of a MOSFET. (After Kahng and Atalla, Ref. 4.)

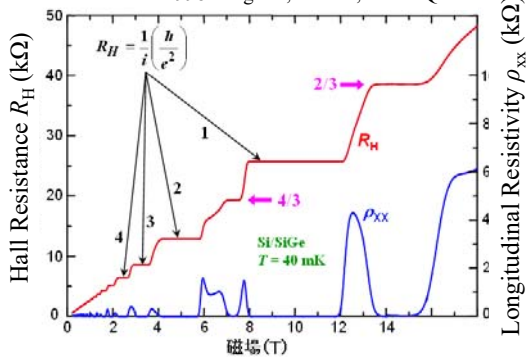


量子ホール効果 Quantum Hall Effect

Nobel prizes
1985 von Klitzing IQHE
1998 Laughlin, Storner, Tsui FQHE

Okamoto Lab.

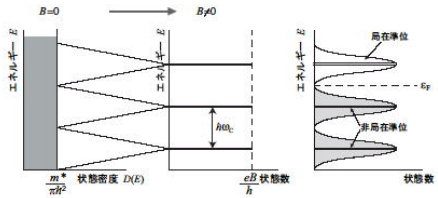




$$E = \frac{\hbar^2 k^2}{2m^*}$$ Landau levels

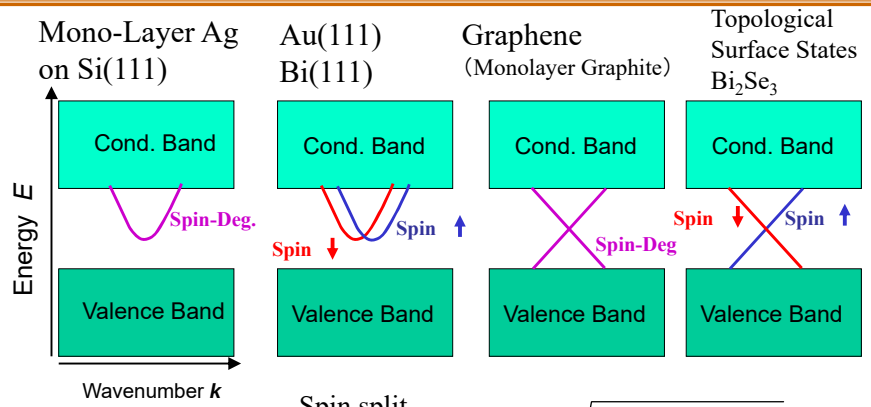
$$E = \hbar\omega_c\left(n+\frac{1}{2}\right) \quad (n = 0,1,2,3...)$$

Cyclotron freq. $\omega_c = \frac{eB}{m^*}$



- Hall resistance is quantized.
  Klitzing constant R_K
  $h/e^2 = 25812.807557(18)\ \Omega$
- Longitudinal resistance is zero.
  ←edge conduction
  ← Topology (Berry's phase)

Various Surface States



$$E = \frac{p^2}{2m^*} = \frac{\hbar^2 k^2}{2m^*}$$

Free-Electron-like
(Non-relativistic)

Spin split
Due to Rashba effect

$$E = \sqrt{(mc^2)^2 + (pc)^2}$$

$$m = 0$$

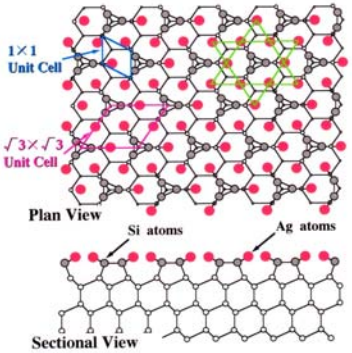
$$E = \pm pc = \pm c\hbar k$$

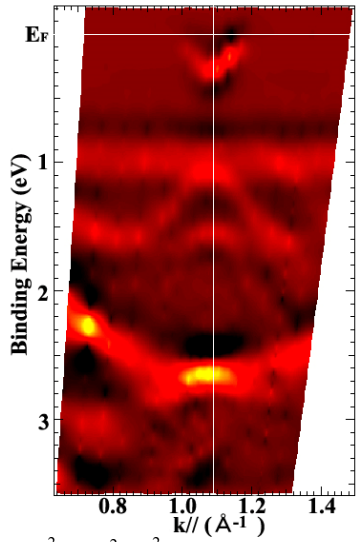
Relativistic

Massless Dirac Electrons

Mono-Layer Ag on Si : Si (111)-√3×√3-Ag Surface

2D Metal
(Monatomic-Layer Metal)



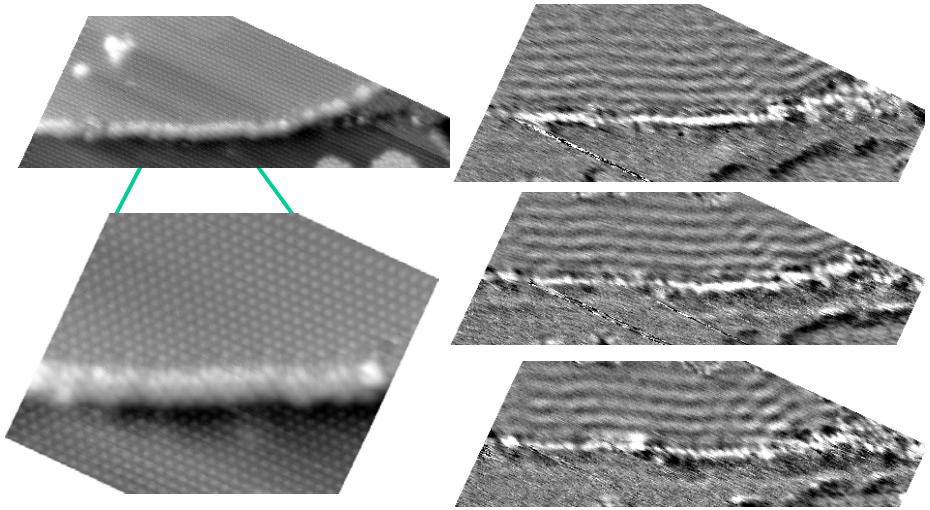


-Inert and atomically flat surface
-Free-electron-like surface state

$$E = \frac{p_{//}^2}{2m^*} = \frac{\hbar^2 k_{//}^2}{2m^*}$$

Standing Waves on Si(111)-√3×√3-Ag Surface at 65K

STM Images **dI/dV Images**

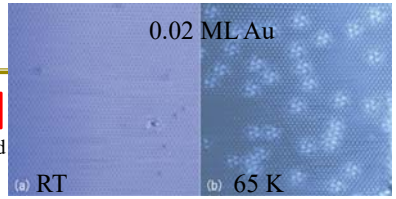


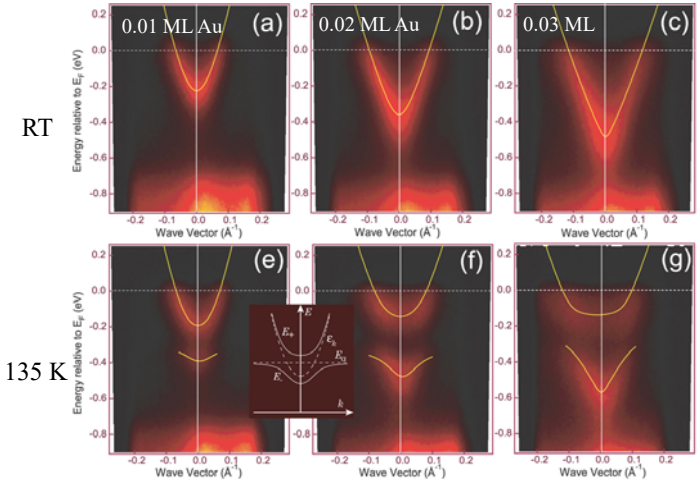
Carrier Doping Into Surface States to Change Electrical Conductivity

Au Adsorption on Si(111)-√3×√3-Ag

1. Carrier doping in the surface-state band
   ⇒ Increase in band occupation
2. Hybridization of the localized state and surface-state band
   ⇒ Band splitting

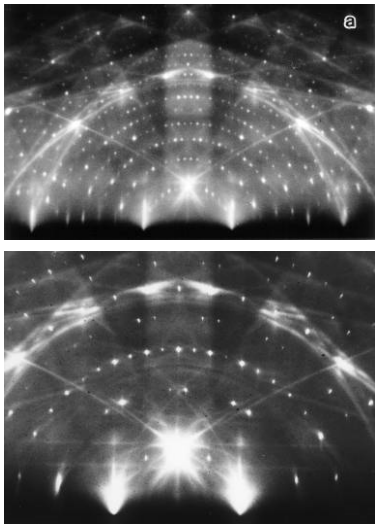
$$\sigma = e \cdot \mu \cdot n$$

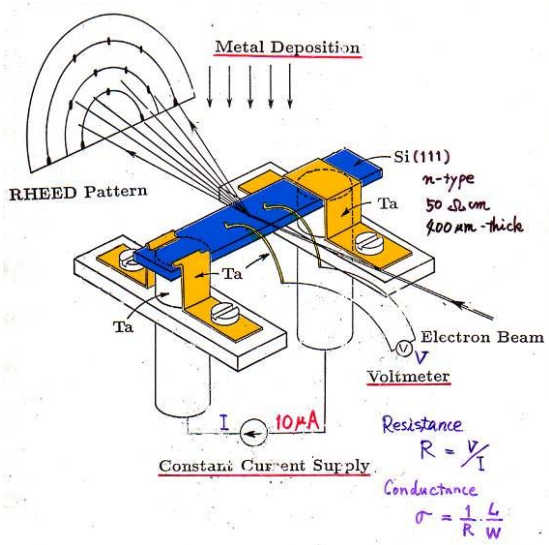




C. Liu, I. Matsuda, R. Hobara, and S. Hasegawa, Phys. Rev. Lett. **96**, 036803 (2006).

Macro-Four-Terminal Measurements in UHV

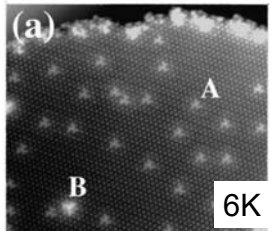


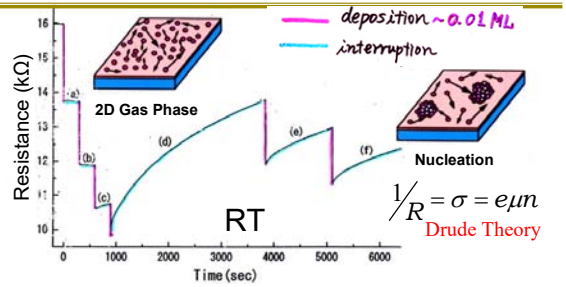


S. Hasegawa, et al., Phys. Rev. Lett. **68**, 1192 (1992)

Carrier Doping into Surface-State Band by Adatoms

Y. Nakajima, *et al.*,
PRB **54**, 14 134(1996); **56**, 6782 (1997).

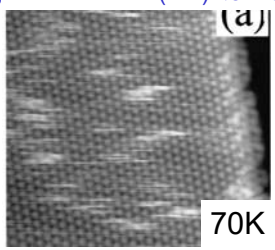




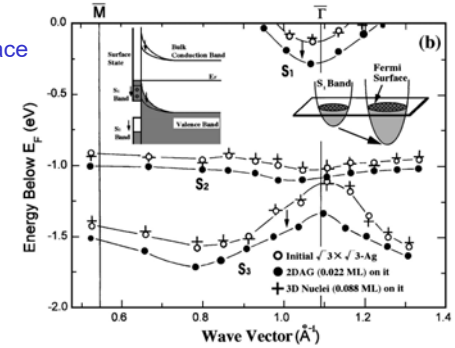
$$1/R = \sigma = e\mu n$$

Drude Theory

Ag Adatoms on Si(111)-√3×√3-Ag Surface

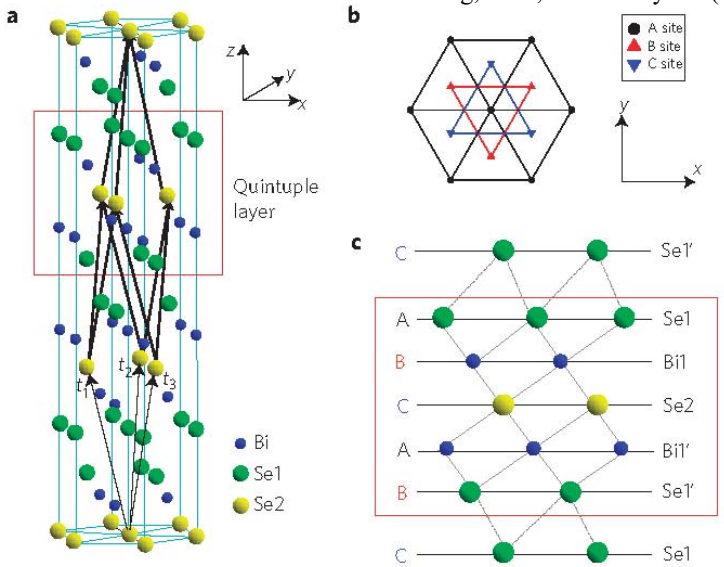


N. Sato, *et al.*, PRB **60**, 16 083(1999).



Crystal Structure of Bi_2Se_3 : Topological Insulator

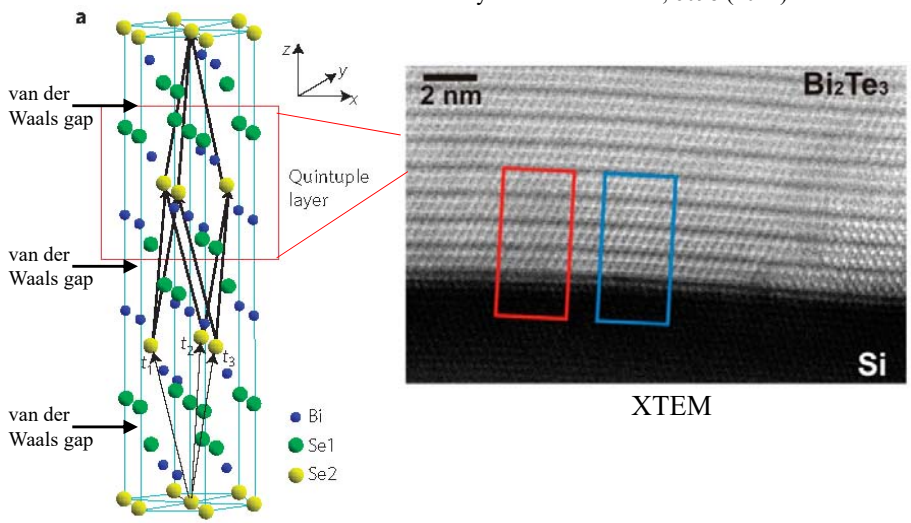
H. Zhang, et al., Nature Physics (May 2009)



Crystal Structure of Bi_2Se_3 (Bi_2Te_3)

H. Zhang, *et al.*, Nature Physics (2009)

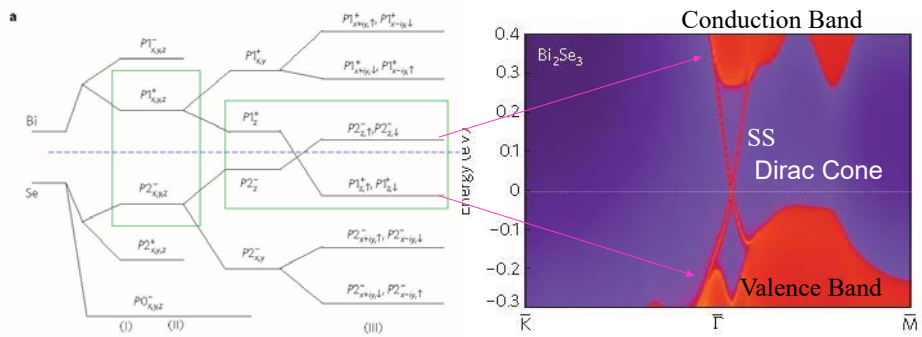
S. Borisova, *et al.*, Cryst. Growth Des. **12**, 6098 (2012)



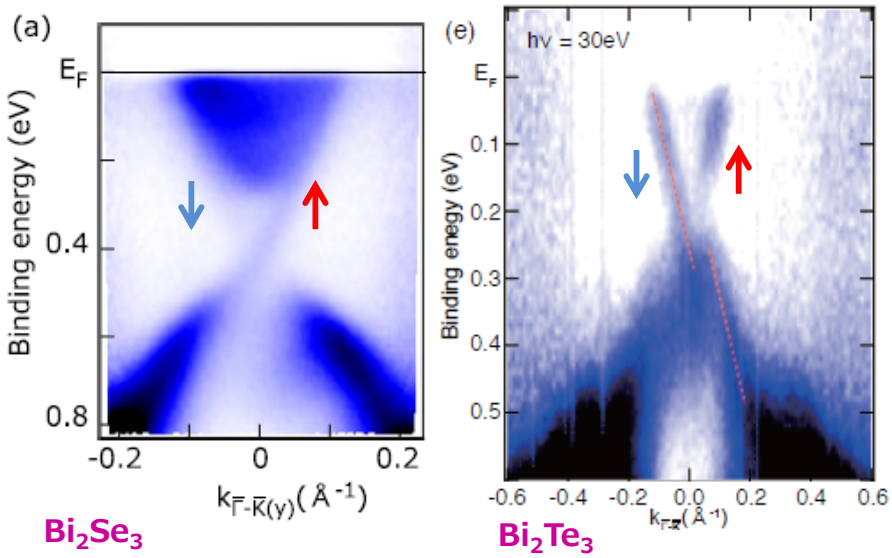
XTEM

Electronic States of Bi_2Se_3 (Theory)

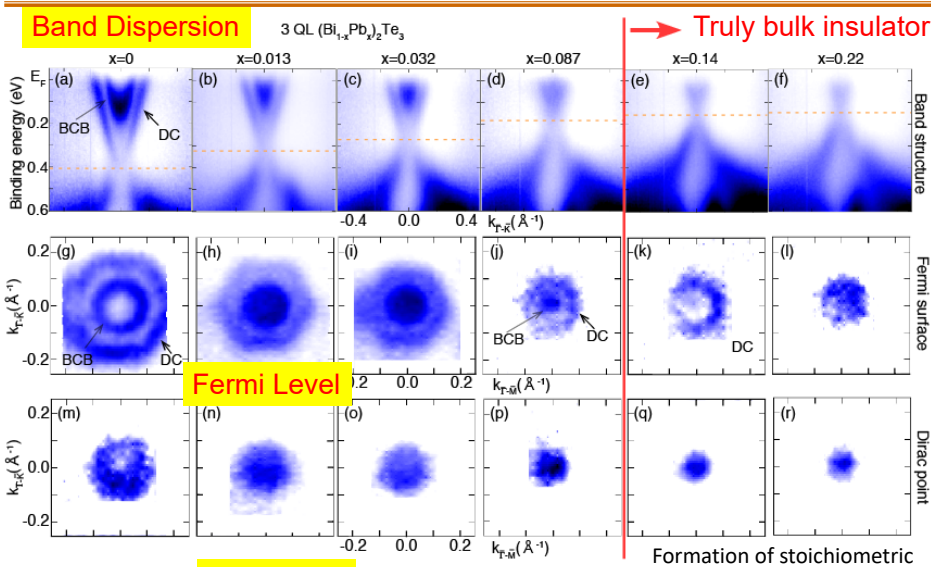
H. Zhang, et al., Nature Physics (May 2009)



Dirac Cones of Topological Insulators



Hole-doping by Pb alloying in Bi_2Te_3

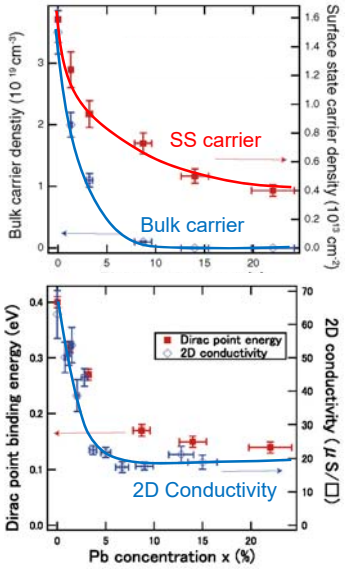


M. Aitani, *et al.*, Jap. J. Appl. Phys.**52** (2013) 110112

Formation of stoichiometric $PbBi_4Te_7$, $Bi_2Te_3(PbTe)_{0.5}$ or Te-Bi-Te-Pb-Te-Bi-Te?

Conductivity of a single Dirac-cone surface state

M. Aitani, et al., Jpn. J. Appl. Phys. **52**, 110112 (2013)



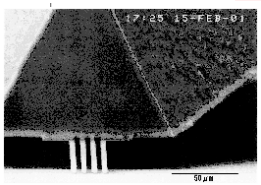
$$\sigma_{SS} \approx 20\mu S \approx 0.5\frac{e^2}{h}$$

Conductivity of
Single (Spin-split) Dirac Cone

Drude Model $\sigma = e\mu n$

Carrier Density $n = 4\times10^{12}$ cm^{-2}

$\Rightarrow$ Mobility $\mu = 30$ cm^2/Vs



Not high mobility!

Four-Point Probe Method for Transport Measurements

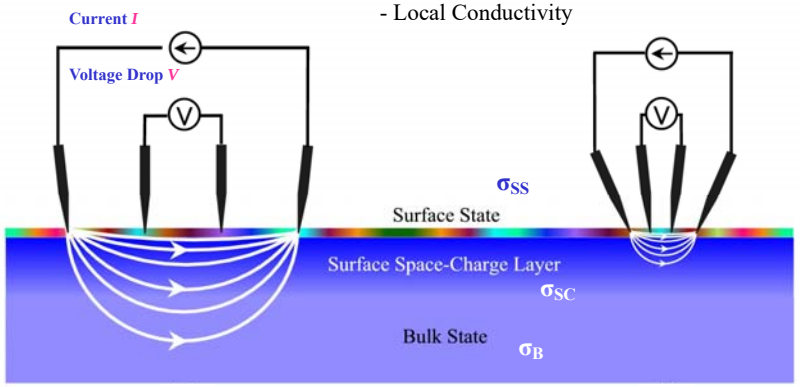
Electrical Resistance

$$R = \frac{V}{I}\cdot C$$

C: Correction Factor

- Contact Resistance
- Three Parallel Conduction Channels
- Surface Sensitivity
- Local Conductivity

$\sigma_{meas} = \sigma_{SS} + \sigma_{SC} + \sigma_B$



Macro-4-Point Probe

Bulk-Sensitive
(Surface -Insensitive)

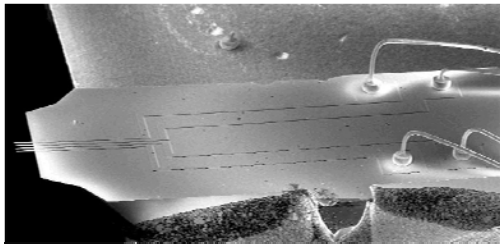
Micro-4-Point Probe

Surface -Sensitive

Temperature-Variable Monolithic Micro-Four-Point Probe

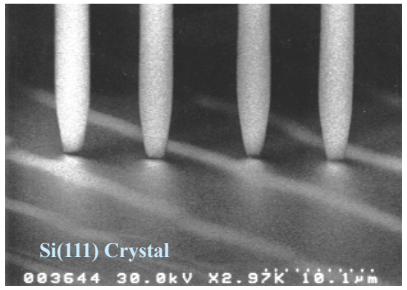
I. Shiraki, et al., Surf. Rev. Lett. **7** (2000) 533.
C. L. Peteresen, et al., Appl. Phys. Lett. **77** (2000) 3782.
S. Hasegawa, et al., J. Phys: Cond. Matters **14** (2002) 8379.
T. Tanikawa, *et al.*, e-J. Surf. Sci. Nanotech. **1** (2003) 50.

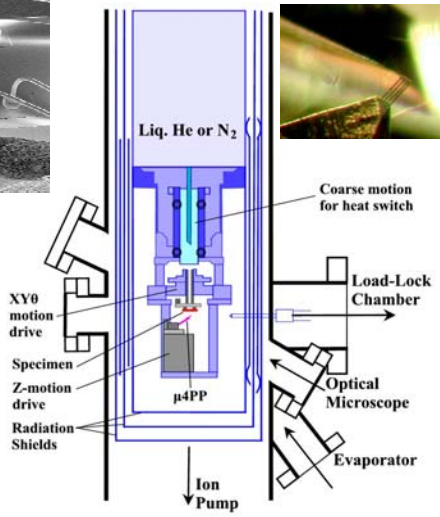
Developed at Denmark Technical University



Commercially available;
http://www.capres.com

Contacting to Si(111) sample surface in SEM





1D, 2D, and 3D conduction

$3D: R \propto d^{-1}$

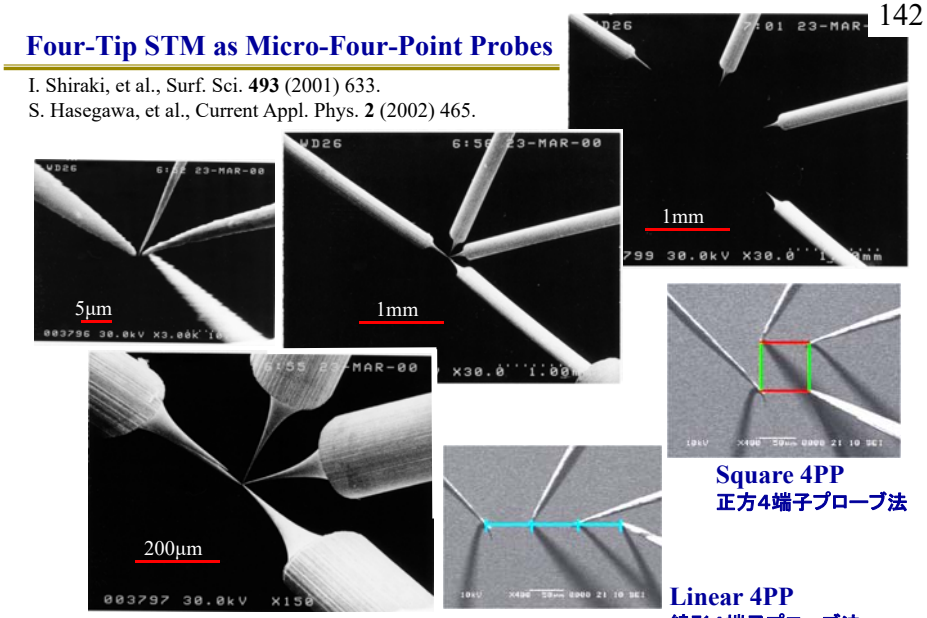
$2D: R \propto d^{0}$

$1D: R \propto d$

d: probe distance (length of measured area of sample)

Four-Tip STM as Micro-Four-Point Probes

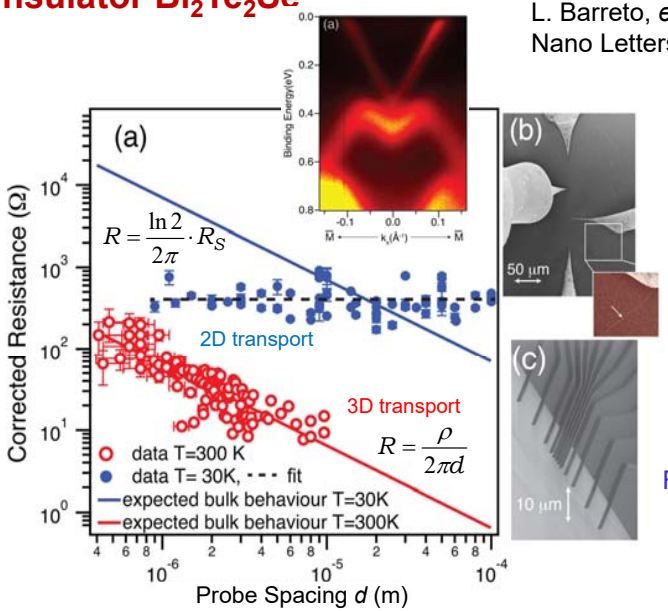
I. Shiraki, et al., Surf. Sci. **493** (2001) 633.
S. Hasegawa, et al., Current Appl. Phys. **2** (2002) 465.



SEM Images of Independently Driven Four W-Tips in UHV

Surface-Dominated Transport on a Bulk Topological Insulator Bi_2Te_2Se

L. Barreto, *et al.*,
Nano Letters **14**, 3755 (2014)

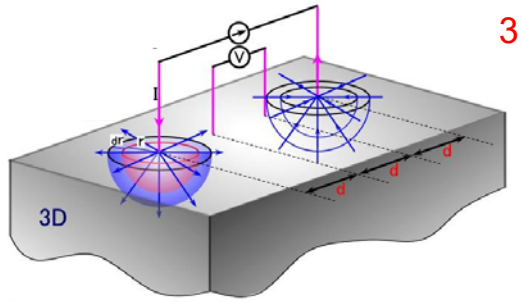


LT: Bulk carriers are frozen out.
⇒ SS transport only

⇒ Mobility
$\mu = 390 \mathrm{cm}^2/\mathrm{Vs}$

Fairly high mobility!

Current Distribution vs. Probe-Spacing Dependence of Resistance



3D transport

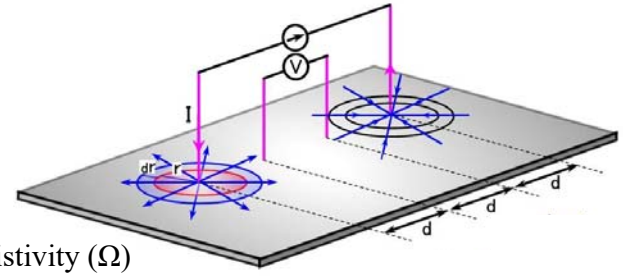
$$R = \frac{V}{I} = \frac{\rho}{2\pi d}$$

ρ : 3D Resistivity (Ωcm

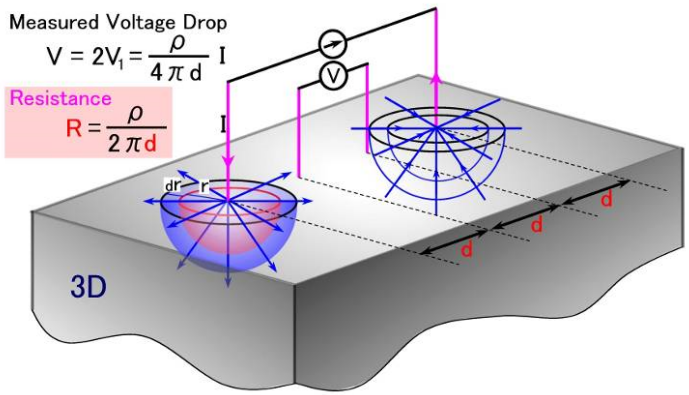
2D transport

$$R = \frac{V}{I} = \frac{\ln 2}{2\pi} \cdot R_S$$

R_S: Sheet Resistivity (Ω)



Four-Point Probe Resistance for a 3D sample



Potential Difference between the two hemispheres

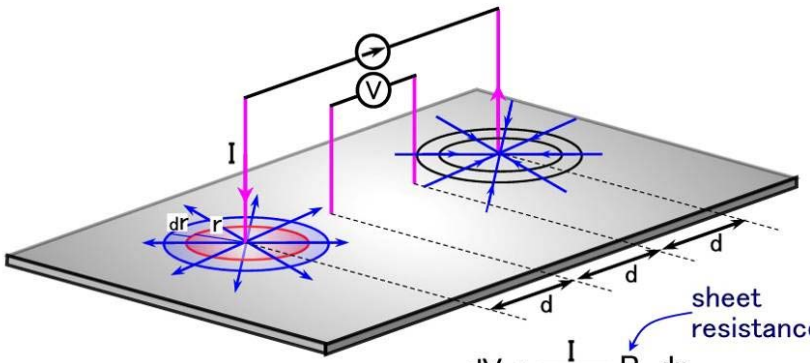
$$dV_1 = \frac{I}{2\pi r^2}\rho\, dr$$

resistivity [Ω cm]

Voltage Drop due to Current Injected = Voltage Drop due to Current Flow Out

$$V_1 = \int_d^{2d} \frac{I}{2\pi r^2}\rho\, dr = \frac{\rho}{4\pi d} I$$

Four-Point Probe Resistance for a 2D sample



sheet resistance [Ω]

$$dV_1 = \frac{I}{2\pi r} R_S\, dr$$

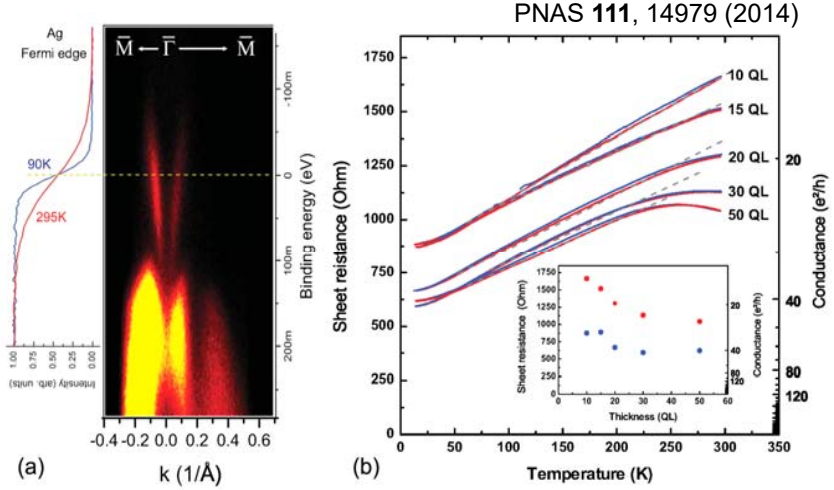
$$V = 2V_1 = \frac{\ln 2}{\pi} R_S I$$

$$V_1 = \int_d^{2d} \frac{I}{2\pi r} R_S\, dr = \frac{R_S I}{2\pi}\cdot \ln\frac{2\cancel{d}}{\cancel{d}} = \frac{\ln 2}{2\pi} R_S I$$

$R = \frac{\ln 2}{2\pi} R_S$: independet of d

Intrinsic conduction through topological surface states of insulating Bi_2Te_3 epitaxial thin films on BaF_2 (111)

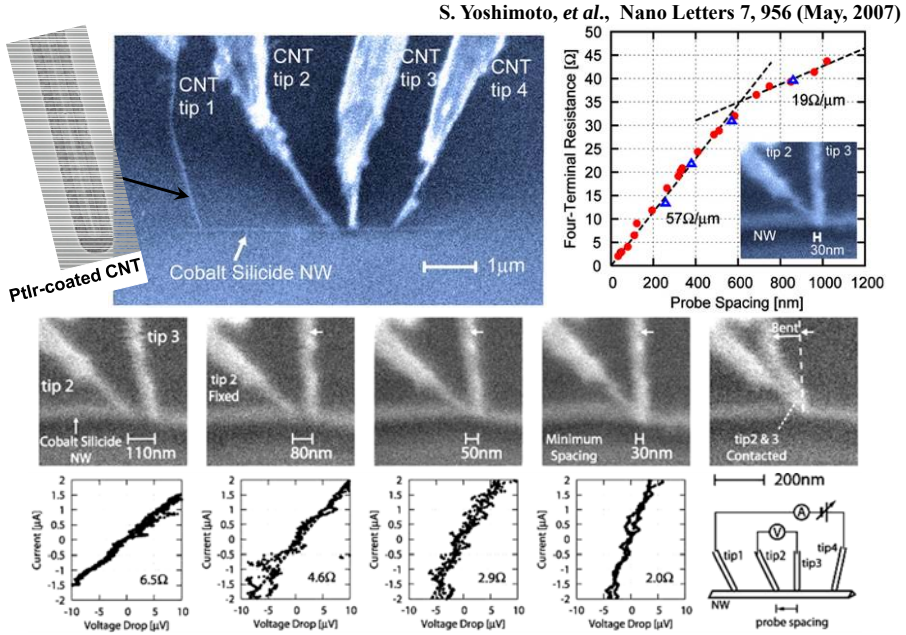
K. Hoefer, *et al.*,
PNAS **111**, 14979 (2014)



⇒ Mobility $\mu = 4{,}600\ \text{cm}^2/\text{Vs}$ Very high mobility!

Four-Tip STM with 20 nm Tip Spacing

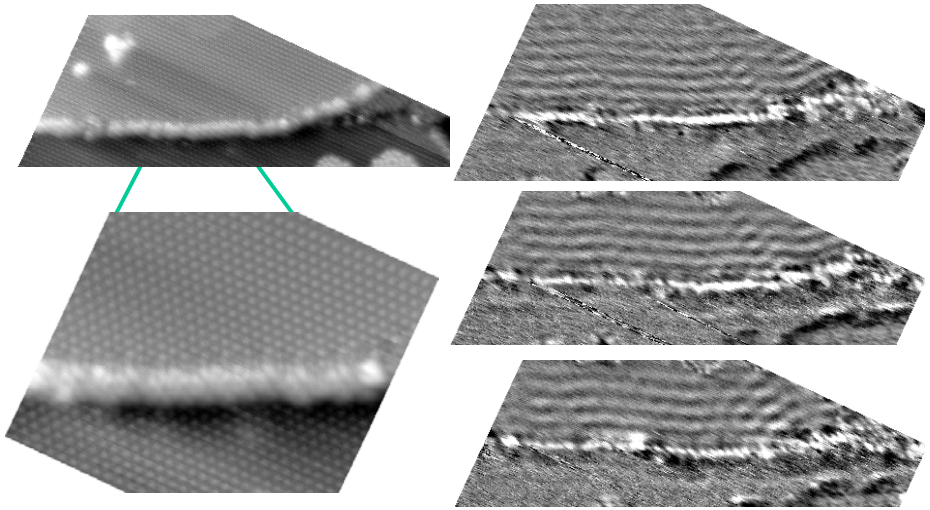
S. Yoshimoto, *et al.*, Nano Letters 7, 956 (May, 2007)



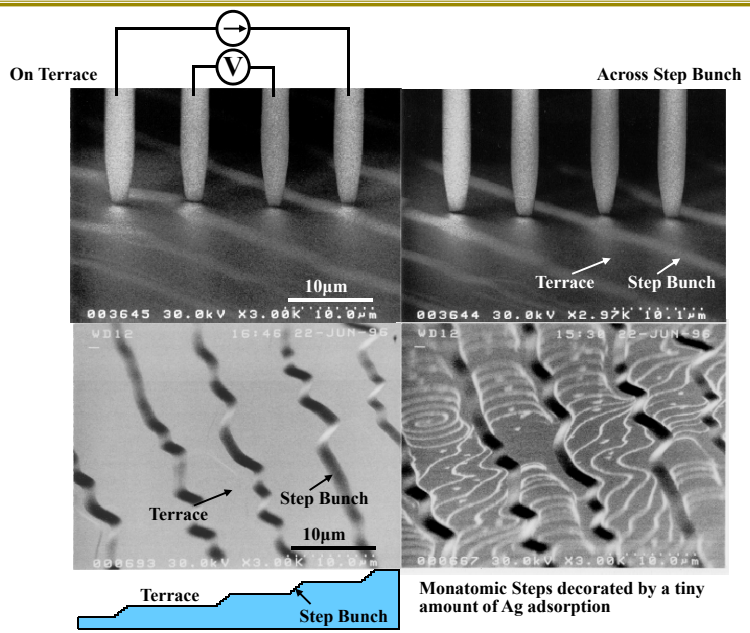
Electrical Resistance due to an Atomic Step

Standing Waves on Si(111)-√3×√3-Ag Surface at 65K

STM Images **dI/dV Images**



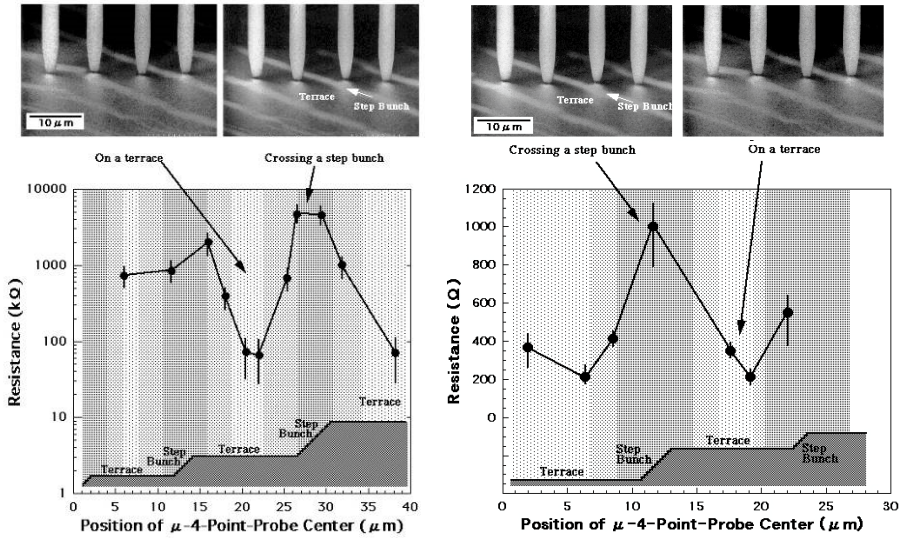
Resistance Measurements Across a Step Bunch or On a Terrace



Monatomic Steps decorated by a tiny amount of Ag adsorption

Line Profile of Resistance measured across Step Bunches at RT in UHV

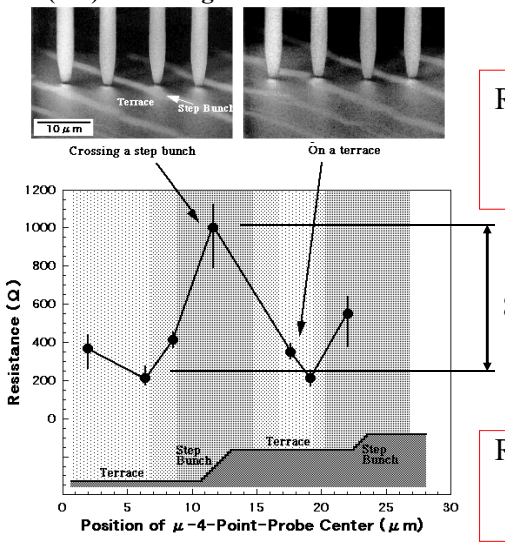
Si(111)-7×7 Clean Surface **Si(111)-√3×3-Ag Surface**



Line Profile of Resistance measured through Step Bunches at RT in UHV

S. Hasegawa et al.,J. Phys.: Cond. Matter **14**, 8379 (2002).

Si(111)-√3×3-Ag Surface



R= 800Ω/300 steps ～3Ω
Resistance of a single step
(Experiment)

800Ω

Good Match

R=1/G～5Ω
Resistance of a single step
(Friedel振動から)

Tunneling Transport —Landauer Formula (Tsu-Esaki) —

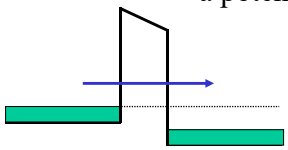
I. Matsuda, *et al.*, Physical Review Letters **93**, 236801 (Dec, 2004)

1D: $G = \frac{2e^2}{h} \cdot T(\mu)$ $[\Omega^{-1}]$

2D: $g = \frac{2e^2}{h} \frac{k_F}{\pi} \cdot T(\mu)$ $[\Omega^{-1}m^{-1}]$

3D: $g = \frac{2e^2}{h} \frac{k_F^2}{4\pi} \cdot T(\mu)$ $[\Omega^{-1}m^{-2}]$

T(μ): Transmission Probability through
a potential barrier/a inlet・exit



ナノワイヤにおける電気伝導 Electrical Conduction through a nano-wire

谷城（東工大）

通常のワイヤ $l >> l_{mfp}$

l

w

ℓ_{mfp}: Mean free path, 平均自由工程

拡散的電子伝導
diffusive electron transport

⇒ Ohm's Law $G \propto \frac{w}{\rho \cdot \ell}$

ナノワイヤ $l < l_{mfp}$

w

弾道的電子伝導
ballistic electron transport

長さに依存しないコンダクタンス

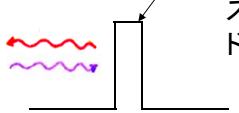
λ_f: Fermi wave length , n: number of modes

$w \approx \lambda_f \rightarrow G = nG_0 = n\frac{2e^2}{h}$ （量子化）

⇒ independent of Length ℓ

Conductance Quantization

電子定在波



ポテンシャル障壁
ステップ
ドメイン境界

ρ(E, x)（局所状態密度）
$\propto |\Psi_i+\Psi_r|^2$
$\propto \{1+|R|^2+2|R|\cos(2k_x \cdot x-\eta)\} \cdot |u(x,y)|^2$

定在波　原子像

2次元　ブロッホ波

入射波 $\Psi i=\exp[i(k_x \cdot x+k_y \cdot y)] \cdot u(x, y)$

$E=E_0+\frac{\hbar^2}{2m^*}(k_x^2+k_y^2)$

u(x,y)= cell function

定在波の波長$\lambda = \frac{\pi}{k_x}$

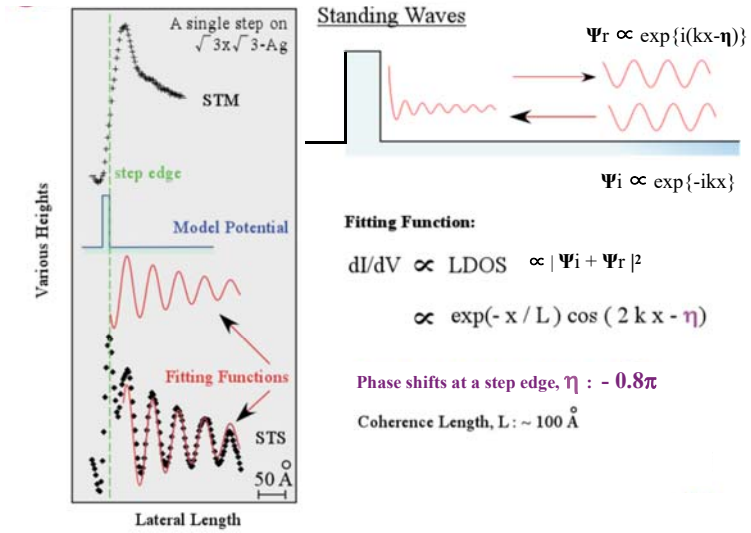
反射波 $\Psi_r=R \cdot \exp[i(-k_x \cdot x+k_y \cdot y)] \cdot u(x, y)$

$R=|R| \cdot \exp[i\eta]$

η: 反射位相シフト

Phase Shift in Reflection at a Step Edge

I. Matsuda, *et al.*, Physical Review Letters **93**, 236801 (Dec, 2004)

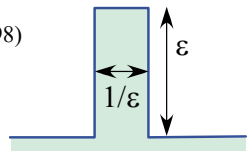


Phase Shift, Transmission Coefficient, and Conductance

Transmission through δ- potential

L. Buergi, et al. Phys. Rev. Lett. **81**, 5370 (1998)

Transmission Prob. $T = 1 - \dfrac{1}{1+\tan^2(\eta+\pi)}$



$\eta = -0.8\pi \Rightarrow T = 0.3 \sim 0.4$

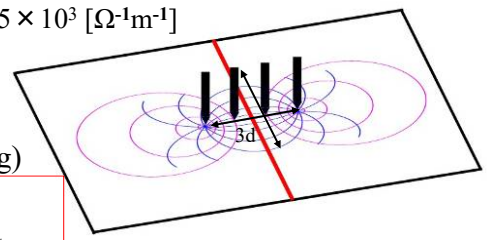
2D Landauer Formula $g = \dfrac{2e^2}{h}\dfrac{k_F}{\pi}\cdot T(\mu)$ $[\Omega^{-1}m^{-1}]$

Measured Conductance $= 5 \times 10^3\ [\Omega^{-1}m^{-1}]$

$G \sim g \cdot 3d$

$k_F = 0.1 Å^{-1}$

$d = 8\mu m$ (Probe Spacing)



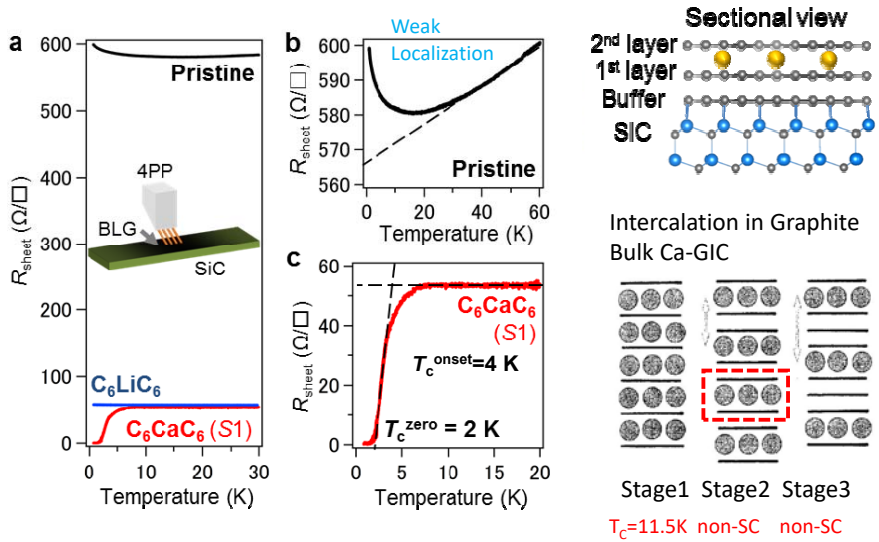
$R = 1/G \sim 5\Omega$

Resistance of a single step

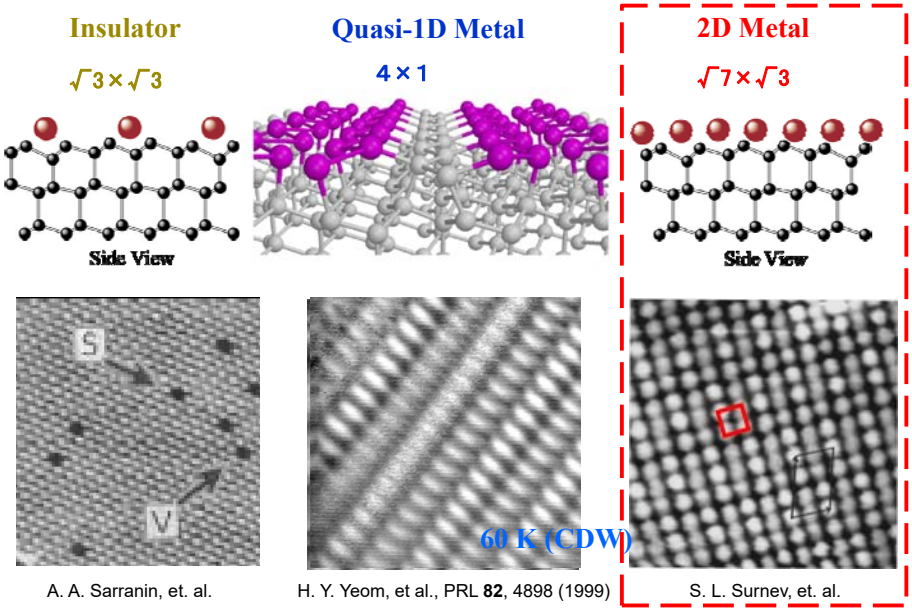
Atomic-Layer Superconductivity
Surface-State Superconductivity

Resistance of Bilayer Graphene w/wo Ca/Li

S. Ichinokura, K. Sugawara, A. Takayama, T. Takahashi, and S. Hasegawa:
"*Superconducting Calcium-Intercalated Bilayer Graphene*" , ACS Nano **10**, 2761-2765 (Jan, 2016)



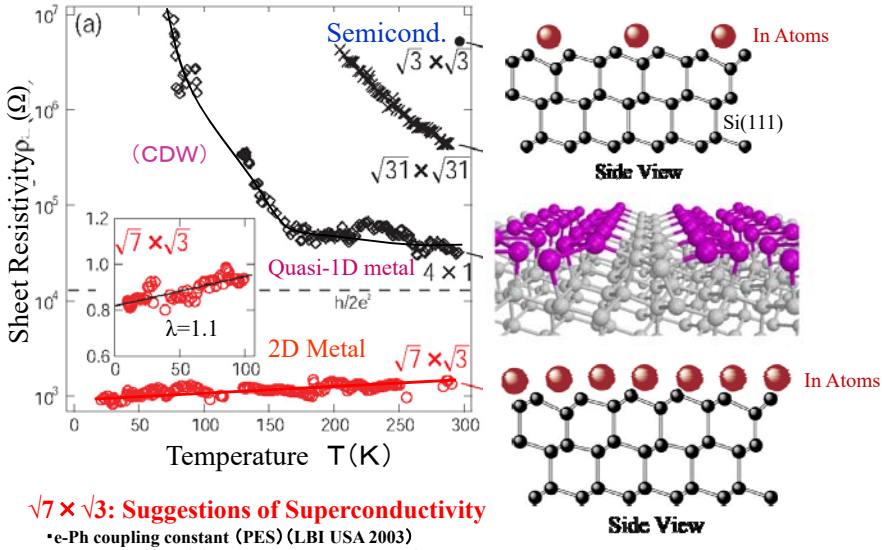
Indium-adsorbed Silicon (111) Surface



A. A. Sarranin, et. al. H. Y. Yeom, et al., PRL **82**, 4898 (1999) S. L. Surnev, et. al.

Electrical Resistance of (Sub)Monolayer In-adsorbed Si

S. Yamazaki, et. al., Phys. Rev. Lett., **106**, 116802 (2011).

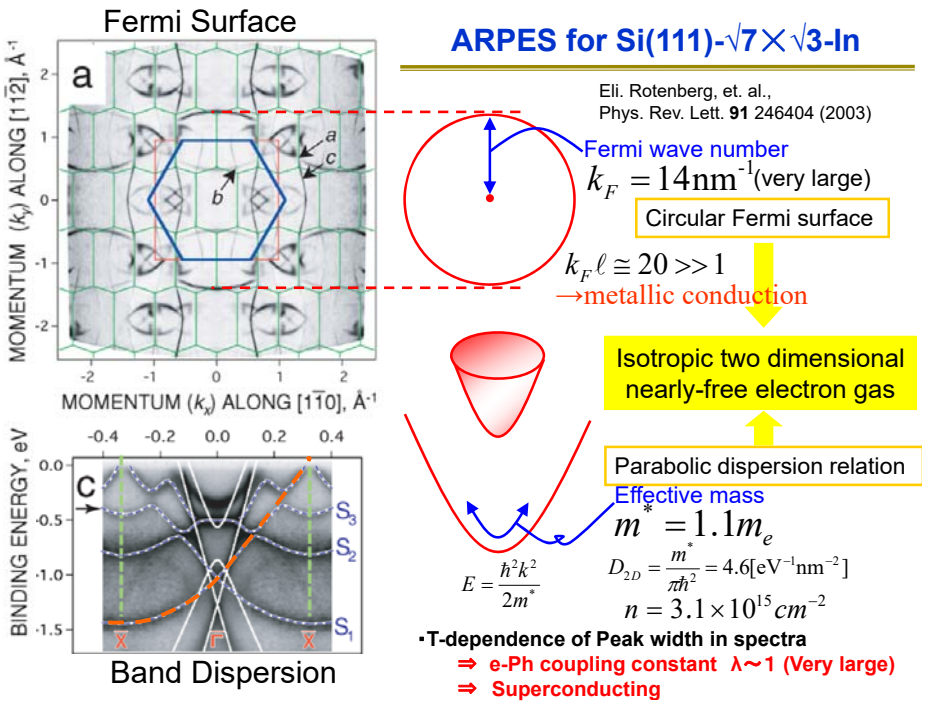


√7×√3: Suggestions of Superconductivity

- e-Ph coupling constant (PES) (LBI USA 2003)
- Energy gap (STS) (Tsinghua Univ. China 2010)

ARPES for Si(111)-√7×√3-In

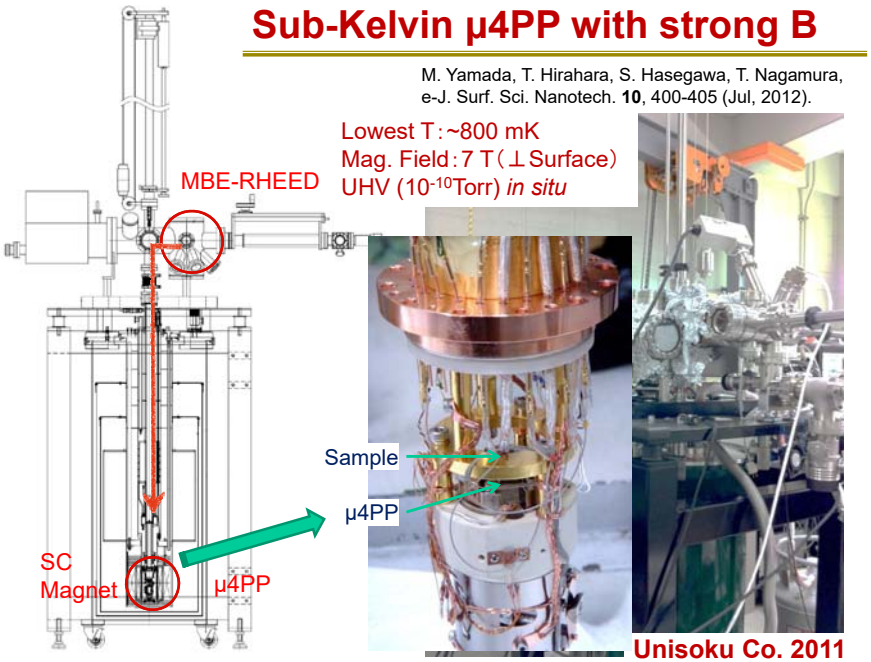
Eli. Rotenberg, et. al., Phys. Rev. Lett. **91** 246404 (2003)



- **T-dependence of Peak width in spectra**
  - ⇒ e-Ph coupling constant λ~1 (Very large)
  - ⇒ Superconducting

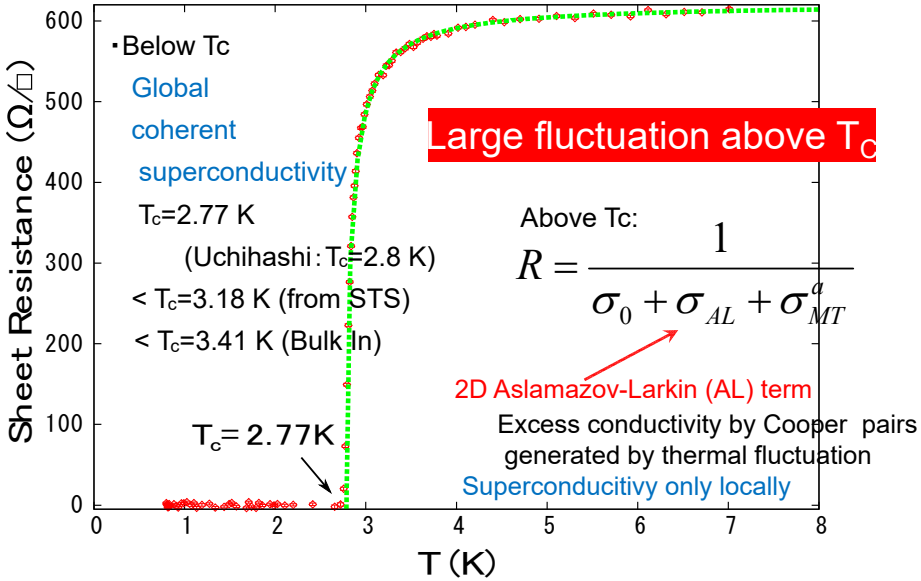
Sub-Kelvin μ4PP with strong B

M. Yamada, T. Hirahara, S. Hasegawa, T. Nagamura, e-J. Surf. Sci. Nanotech. **10**, 400-405 (Jul, 2012).

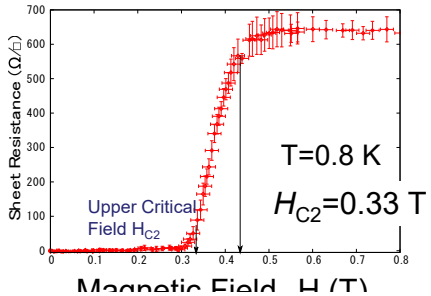


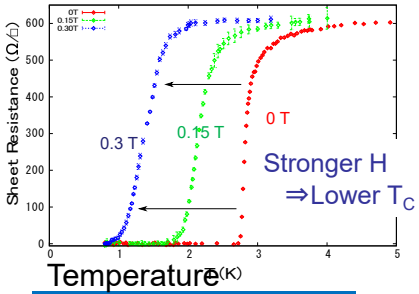
Unisoku Co. 2011

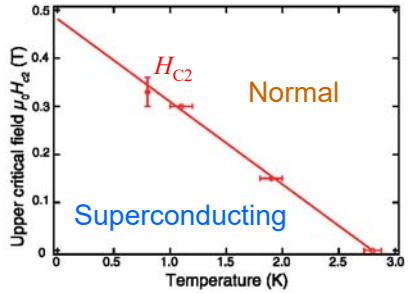
Superconductivity at Si(111) -√7x√3-In



Si(111)-√7x√3-In : Under Magnetic Field







$$H_{C2}(T) = \frac{\phi_0}{2\pi\mu_0\xi(T)^2} = \frac{\hbar}{2e\mu_0\xi(0)^2}\left(1-\frac{T}{T_c}\right)$$

Very high H_c (=short ξ)

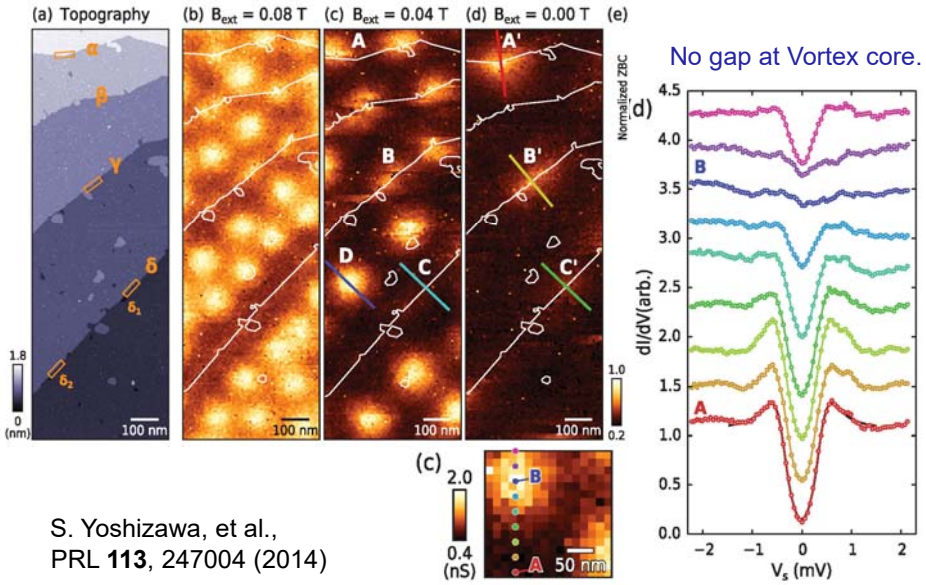
Strong Critical Field $H_{c2}(0)$~5000 Oe

cf. bulk In : H_c=293 Oe

Short Coherence Length

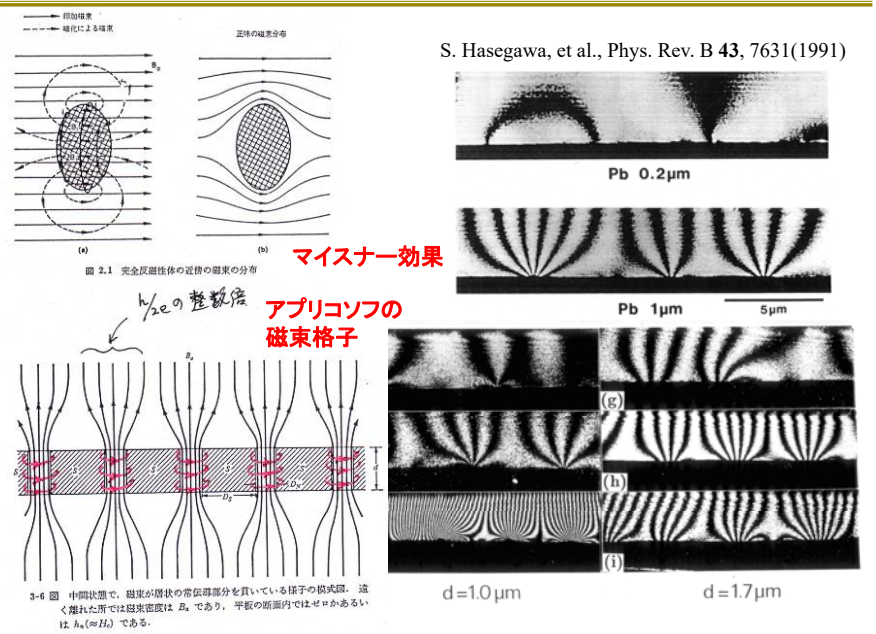
$\xi_{GL}(0)$~25 nm << 330 nm(Bulk In)

Vortex and Scanning Tunneling Spectra of Si(111)-√7 × √3-In Double Layer Superconductor



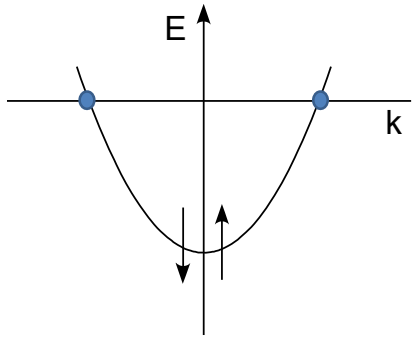
S. Yoshizawa, et al.,
PRL **113**, 247004 (2014)

超伝導薄膜を貫く磁束量子　ー電子ホログラフィによる磁束量子の観察ー



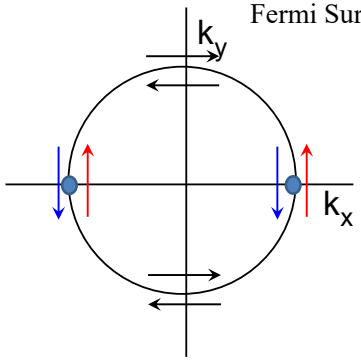
Cooper Pairs in Free-Electron Band

Band Dispersion



$$E = \frac{\hbar^2}{2m}k^2$$

Fermi Surface

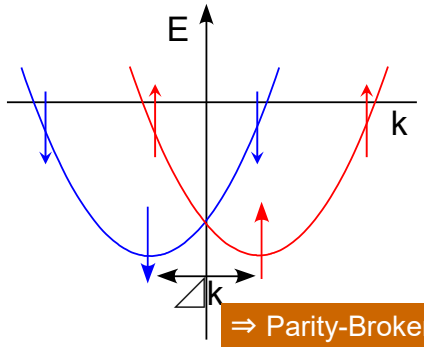


Singlet $|k\uparrow\rangle|-k\downarrow\rangle - |k\downarrow\rangle|-k\uparrow\rangle$

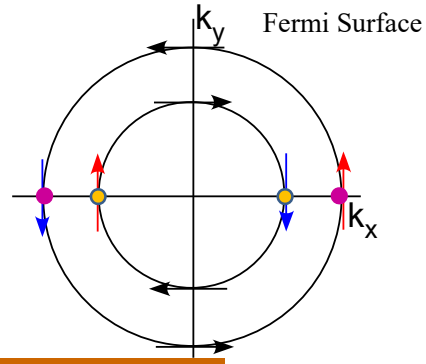
Triplet $|k\uparrow\rangle|-k\uparrow\rangle$, $|k\downarrow\rangle|-k\downarrow\rangle$,
$|k\uparrow\rangle|-k\downarrow\rangle + |k\downarrow\rangle|-k\uparrow\rangle$

Spin Split and Cooper Pairs in Free-Electron Band

Band Dispersion



Fermi Surface



⇒ Parity-Broken Superconductors

●Outer $|k\uparrow\rangle|-k\downarrow\rangle$

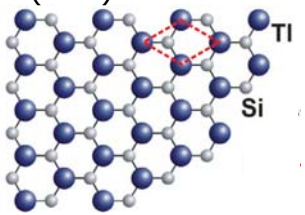
$$|k\uparrow\rangle|-k\downarrow\rangle = \frac{1}{2}\left(|k\uparrow\rangle|-k\downarrow\rangle - |k\downarrow\rangle|-k\uparrow\rangle\right) + \frac{1}{2}\left(|k\uparrow\rangle|-k\downarrow\rangle + |k\downarrow\rangle|-k\uparrow\rangle\right)$$

Singlet (s-wave) Triplet(p-wave)

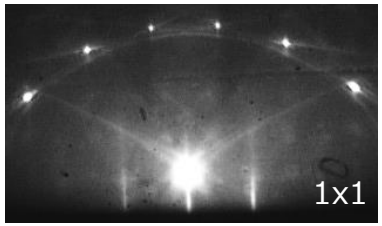
Sample: Si(111)–√3x√3-(Tl, Pb) Surface Superstructure

D. V. Grunev, *et al.*; Sci. Rep. **4,** 4742 (2014).

Si(111)–1x1-Tl

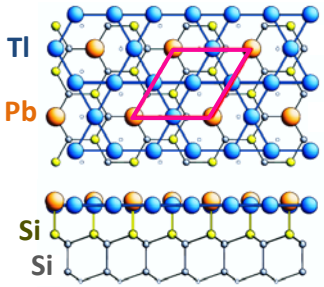


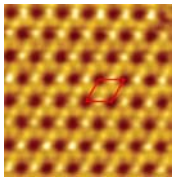
Tl 1ML on 7x7 @ 600K



⇩

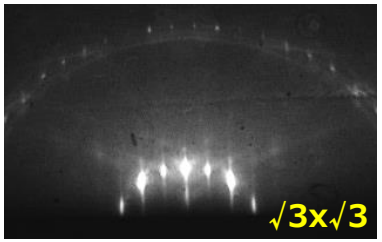
Si(111)–√3x√3-(Tl, Pb)





← Mono-Layer

+Pb 1/3 ML @ 300K

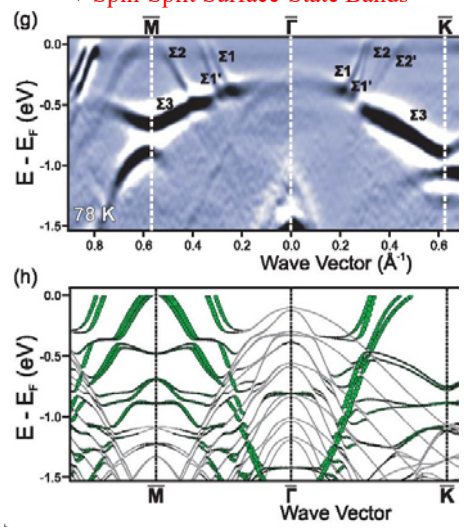


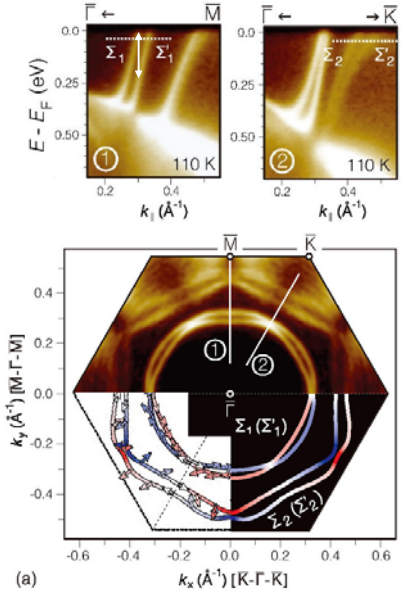
Si(111)-√3 × √3-(Tl, Pb) :Rashba-type SS

D. V. Gruznev, et al., Sci. Rep. **4**, 4742 (2014)

ARPES+Theory

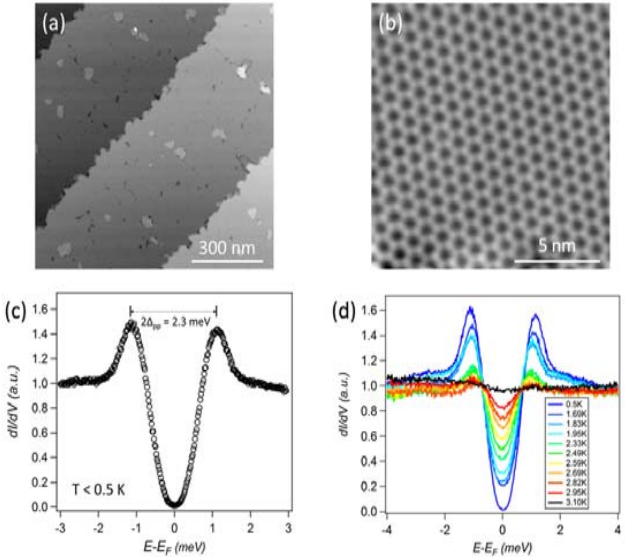
⇒ Spin-Split Surface-State Bands





STS below T_C on Si(111)-√3 × √3-(Tl,Pb) w/o B

T. Nakamura, *et al.*; Phys. Rev. B **98**, 134505 (Oct, 2018).

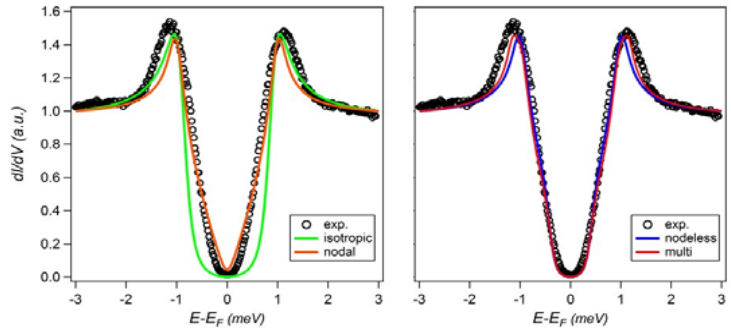


Fitting STS Spectra by Theory

T. Nakamura, *et al.*; Phys. Rev. B **98**, 134505 (Oct, 2018).

Dynes formula : BCS density of states with broadening parameter (Γ)

$$\frac{dI_{ns}}{dV} = \rho_t(0)\rho_n(0)\int_{-\infty}^{\infty} \mathrm{Re}\left\{\frac{|E - i\Gamma|}{\sqrt{((E - i\Gamma)^2 - \Delta^2)}}\right\}\left[\frac{\exp[(E + eV)/k_B T]}{k_B T\{1 + \exp[(E + eV)/k_B T]\}^2}\right] dE$$



s-wave (isotropic gap)

$\Delta = const.$ (1.0 meV)

anisotropic gap

$\Delta = \Delta_0 + \Delta_1 \cos\theta$

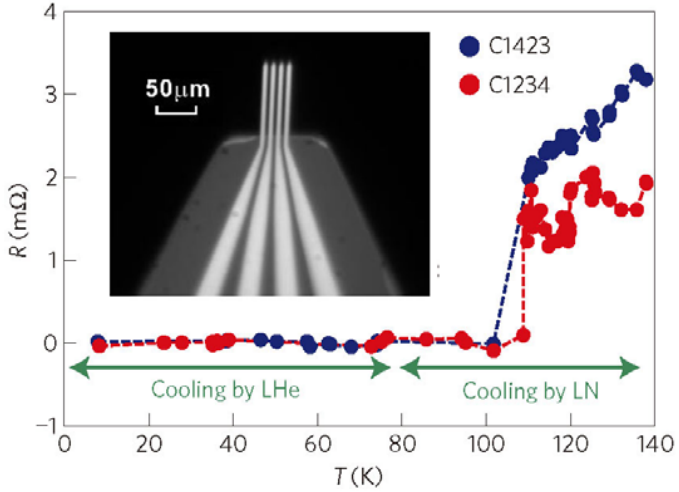
$\Delta_0 = 0.47$ meV

$\Delta_1 = 0.84$ meV

Superconductivity above 100 K in single-layer FeSe films on doped $SrTiO_3$

J-F. Ge, *et al.*, Nature Materials **14**, 285 (Mar, 2015)

measured in situ by μ4PP in UHV



Cf: Bulk FeSe

T_C = 9.4 K

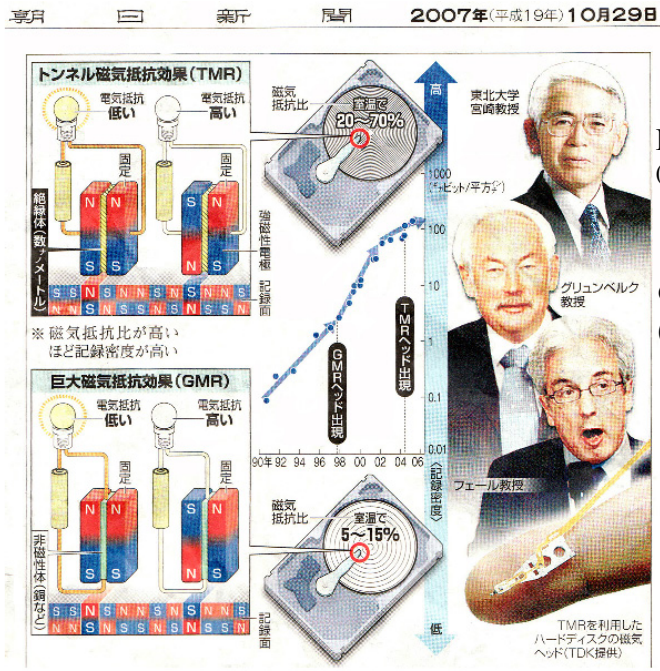
Spin Transport at Surfaces and Edges

Utilizing Spins

- Storage Media
  記憶媒体 Magnetic Disk/Head, MRAM, …

- Energy-Saving /High-Speed Devices
  省エネ・高速素子
  spin current
  spin transistors
  sensors
- 量子情報処理素子
  Quantum Infomration Devices
  Superposition of spin-up and spin-down states

Magnetic Materials
Hetero-, Wire-structures
Diluted Mag. Semiconductors
Magnetic molecues
Graphene
Topological Insulators



Miyazaki
(Tohoku Univ.)

Gruenberg
(Germany)

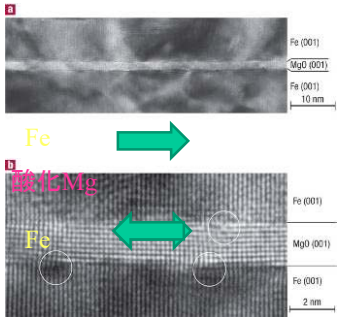
Fert
(France)

The Nobel Prize in Physics 2007

Discovery of Giant MagnetoResistance (GMR) Effect
巨大磁気抵抗効果の発見 ⟶ 磁気ヘッド（ハードディスクの小型化・高密度化）
Magnetic Head (HDs becomes smaller and high-density)

Albert Fert
France
南パリ大学
b. 1938

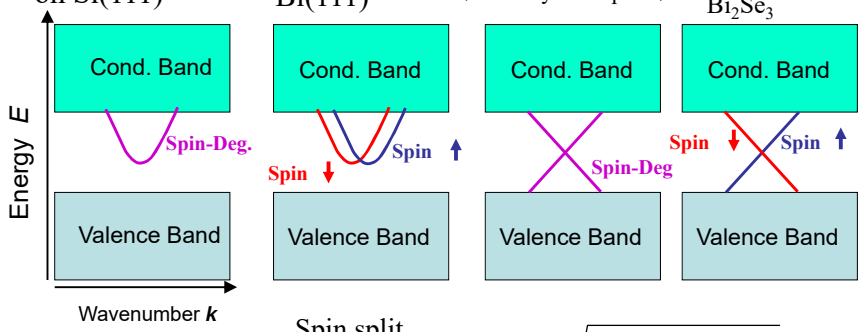
Peter Grünberg
Germany
Julich Inst
b. 1939



S. Yuasa, et al., Nature Materials 3, 868 (2004).

Tunnel MagnetoResistance (TMR) Effect

Various Surface States



$$E = \frac{p^2}{2m^*} = \frac{\hbar^2 k^2}{2m^*}$$

Free-Electron-like
(Non-relativistic)

Spin split
Due to Rashba effect

$$E = \sqrt{(mc^2)^2 + (pc)^2}$$

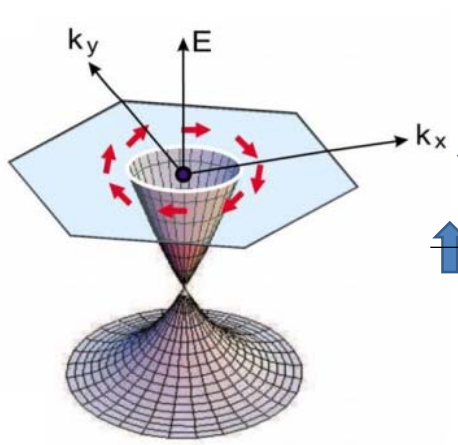
$$m = 0$$

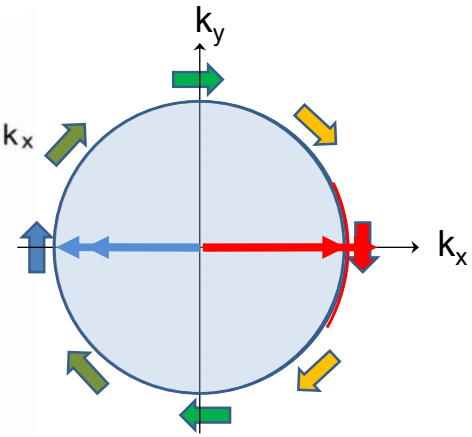
Relativistic

$$E = \pm pc = \pm c\hbar k$$

Massless Dirac Electrons

Spin-Textured Fermi Surface＋Electric Field
⇒Current –Induced Spin Polarization 電流誘起スピン偏極





M. Z. Hasan and C. L. Kane,
Rev. Mod. Phys. **82**, 3045 (2010)

Spin-Textured Fermi Surface
フェルミ面のスピン繊維構造

Spin Hall Effect

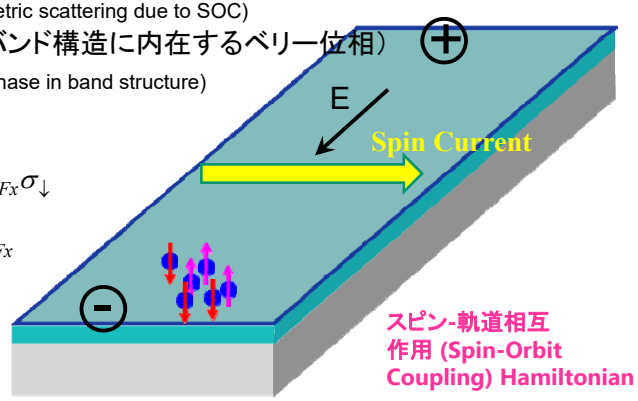
磁場の印加が不要
No external magnetic field
非磁性物質（SOCが強い）
Non-magnetic materials(strong SOC)

・外因性（SOCによる非対称散乱）
Extrinsic reason (asymmetric scattering due to SOC)
・内因性Intrinsic（バンド構造に内在するベリー位相）
Intrinsic reason (Berry phase in band structure)

Spin Current

$$J_S = n_\uparrow \vec{v}_{Fx}\sigma_\uparrow - n_\downarrow \vec{v}_{Fx}\sigma_\downarrow$$

$$\ldots\ldots = \frac{n}{2}\left(\sigma_\uparrow - \sigma_\downarrow\right)\cdot \vec{v}_{Fx}$$

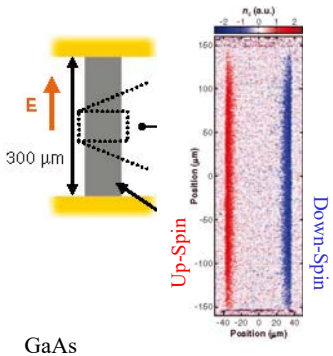


スピン-軌道相互作用 (Spin-Orbit Coupling) Hamiltonian

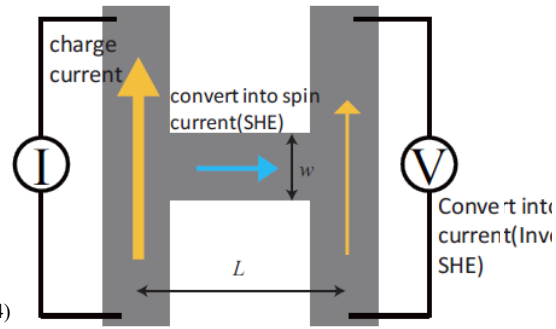
$$H = \frac{1}{2m}p^2 + V(x) + \frac{1}{4mc^2}\sigma\cdot\left(grad\,V\times p\right)$$

Detecting Spin Hall Effect

- Bi_2Se_3; 8QL thick
- Se capping →FIB Fabrication
- Heating → Remove Se Capping



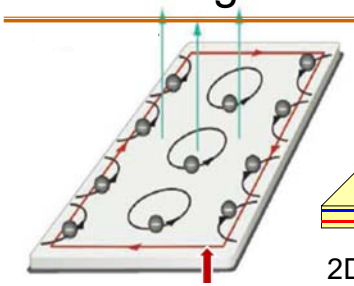
GaAs
Kerr Rotation Microscopy
Y. K. Kato, et al., Science **306**, 1910 (2004)



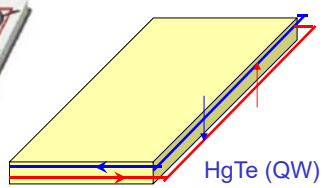
Current → (SHE) Spin Flow → (ISHE) Current/Voltage → Electrical Detection

It should be $L < L_s$ (Spin relaxation length).

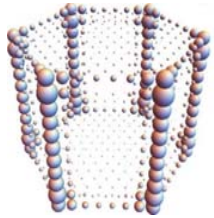
Edge Currents at Q(A)(S)HE



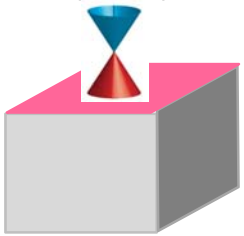
QHE (chiral)



2D Topol. Ins. (QSHE) (helical)



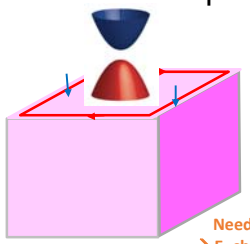
3D Higher-Order Topol. Ins.



3D Topol. Ins.

$E(+k,\uparrow)\neq E(-k,\downarrow)$

Breaking time-reversal symmetry by magnetic order
← Mag. proximity



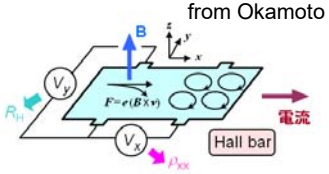
QAHE
1D chiral edge state

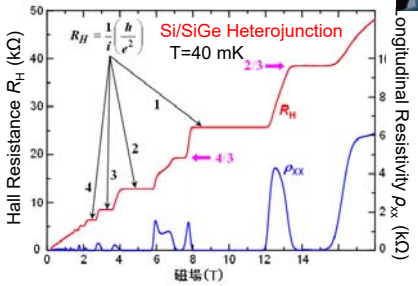
Need E_F tuning
→ E_F should be in the gap.

3D Mag.Topol. Ins. (chiral)

Quantum Hall Effect at 2DEG under strong external mag. field

from Okamoto Lab





- Hall Resistance : quantized in unit of h/e^2
- Longitudinal Resistance : vanishes

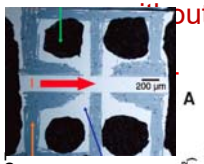
Magnetic Topological Insulator

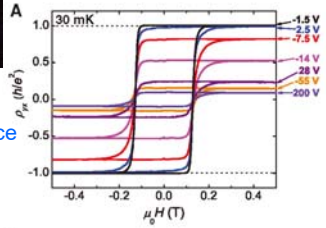
Cr or V doped $Bi_{1-x}Sb_xTe$

without external mag. Field

Anomalous

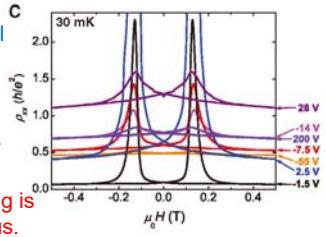
C. Z, Chang, et al., Science 340, 167 (2013)





Hall Resistance Quantized in unit of h/e^2

Longitudinal Resistivity Vanishes

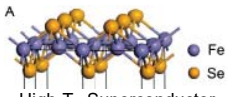


Mag. Proximity to break TRS

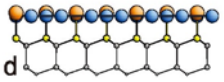
But, bulk doping is Inhomogeneous.

Dissipation-less currents at atomic layers/edges/hinges

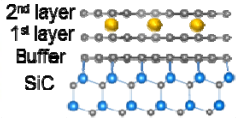
1. Superconducting Current
   - (1) UC FeSe on SrTiO3
   - (2) Monolayer Metals, Pb, In, Ga,…
   - (3) Metal-intercalated Graphene
2. Ballistic Current
3. Edge Currents
   - (1) QHE
   - (2) 2D Topological Insulators (QSHE)
   - (3) 3D Magnetic Topological Ins.(QAHE)
   - (4) Higher-order Topo. Ins.
4. Spin Current
   - (1) Strong Spin-Orbit Int. Mat.(SHE)
   - (2) Heterojunctions at Ferro/Non-mag.



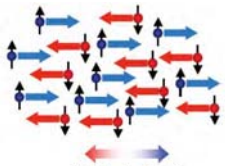
High-T_C Superconductor



Rashba Superconductor

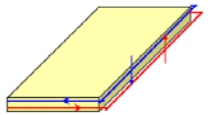


Graphene Superconductor

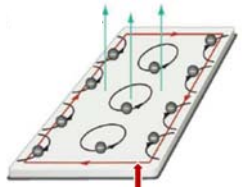


Pure Spin Current.

Hinge states At higher-order Topol.



Edge states at 2D Topol. Ins.



Edge states at QHE