2018年11月12-14日　名古屋大学工学研究科･工学部

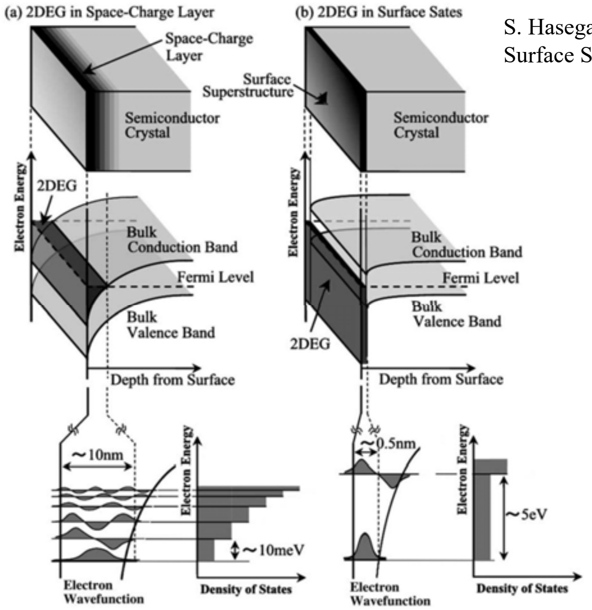
応用物理学特論･応用物理学特別講義（集中講義）

東京大学理学系研究科物理学専攻
長谷川 修司

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

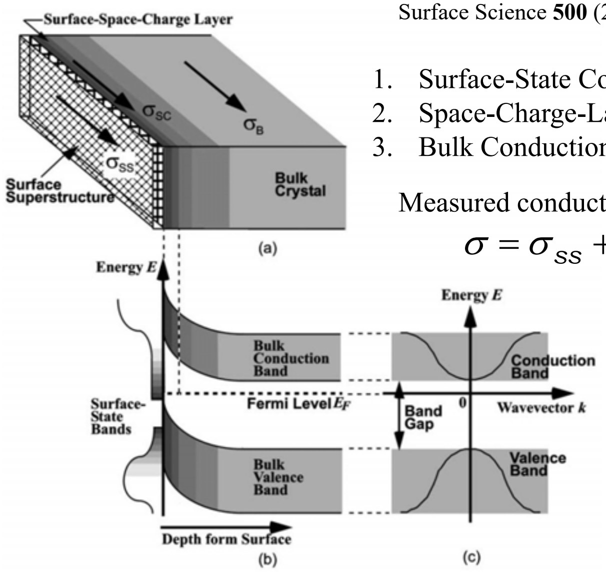
1. Nanoscience and Surface Physics ナノサイエンスと表面物理
   Nanoscience in Nobel Prize
2. Atomic Arrangements at Surfaces 表面原子配列構造
   Scanning Tunneling Microscopy, Electron Diffraction
   走査トンネル顕微鏡、電子回折
3. Surface Electronic States 表面電子状態
   Surface states 表面状態、 Rashba Effect ラシュバ効果
   Topological Surface States 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
   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

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-Control of Band Bending

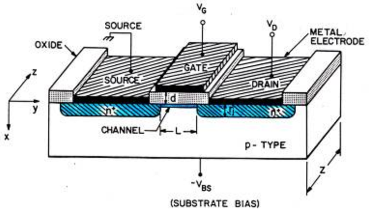
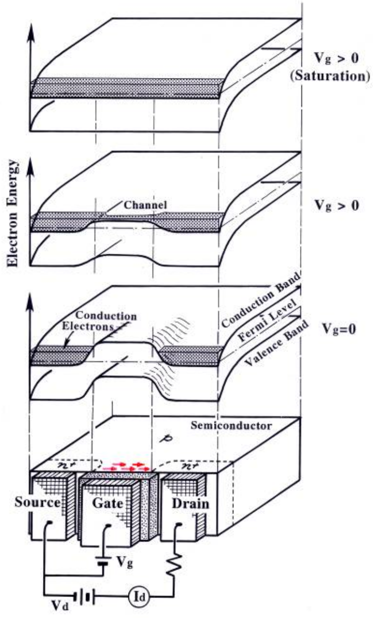


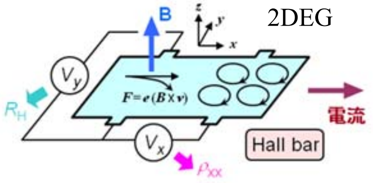
Fig. 3 Schematic diagram of a MOSFET. (After Kahng and Atalla, Ref. 4.)

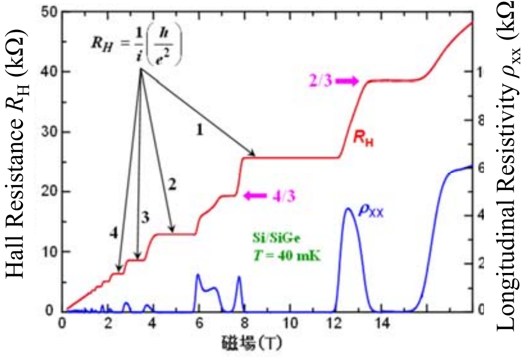


量子ホール効果 Quantum Hall Effect

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

Okamoto Lab.

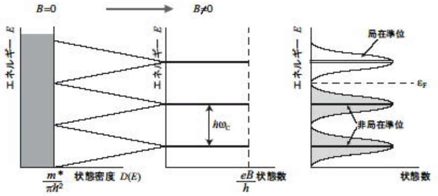




$$E = \frac{\hbar^2 k^2}{2m^*}$$ Landau levels

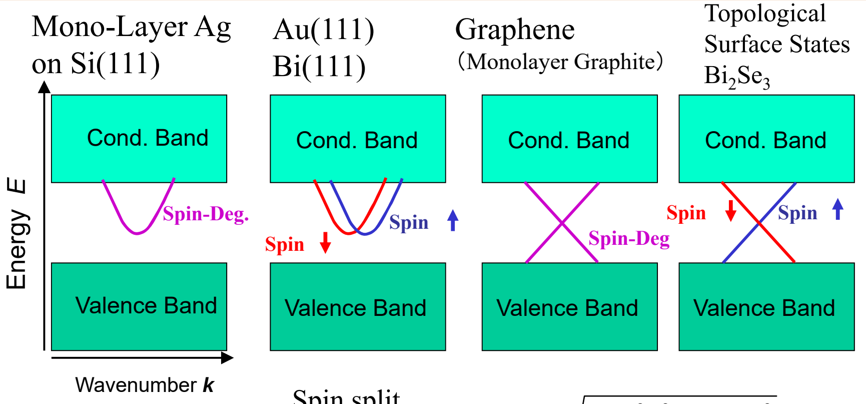
$$E = \hbar\omega_c\left(n + \frac{1}{2}\right) \quad (n = 0,1,2,3\ldots)$$

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



- ▪Hall resistance is quantized.
  Klitzing constant R_K
  $h/e^2 = 25812.807557(18)$ Ω
- ▪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

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

$$\Big\downarrow \; m = 0$$

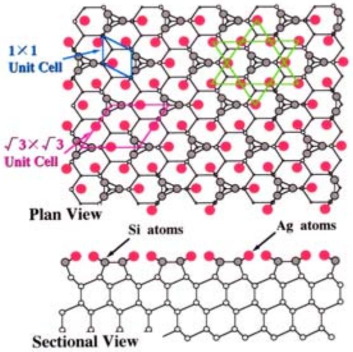
Relativistic

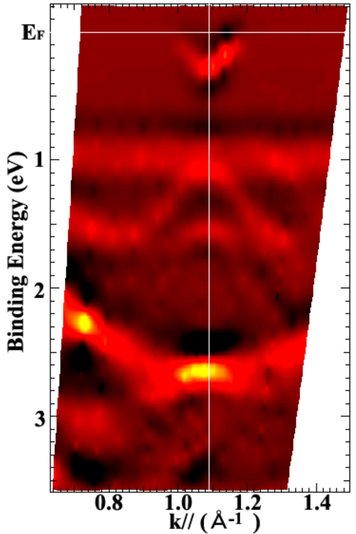
$$E = \pm pc = \pm c\hbar k$$

Massless Dirac Electrons

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

2D Metal
(Monatomic-Layer Metal)



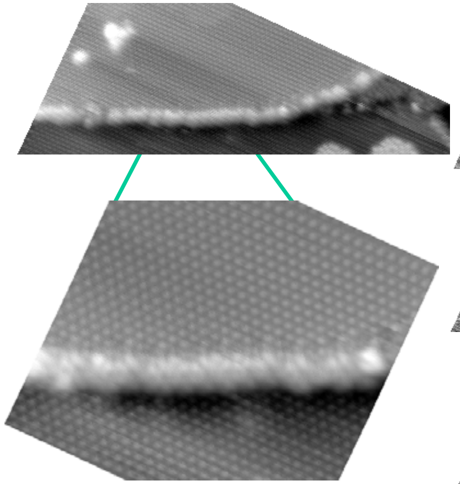


-Inert and atomically flat surface
-Free-electron-like surface state

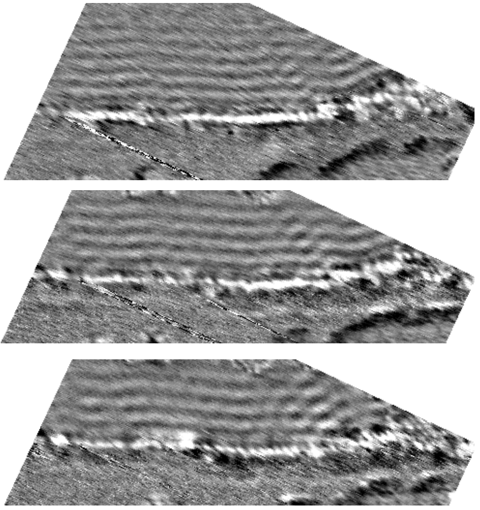
$$E = \frac{p_{//}^2}{2m^*} = \frac{\hbar^2 k_{//}^2}{2m^*}$$

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

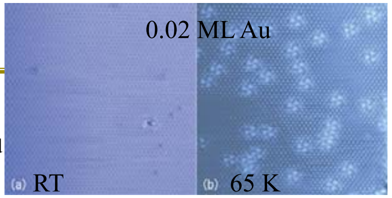
Y. Nakajima, S. Takeda, T. Nagao, S. Hasegawa, and X. Tong:
Surface electrical conduction due to carrier doping into a surface-state band on Si(111)-√3x√3-Ag,
Physical Review B **56** (1997) 6782-6787

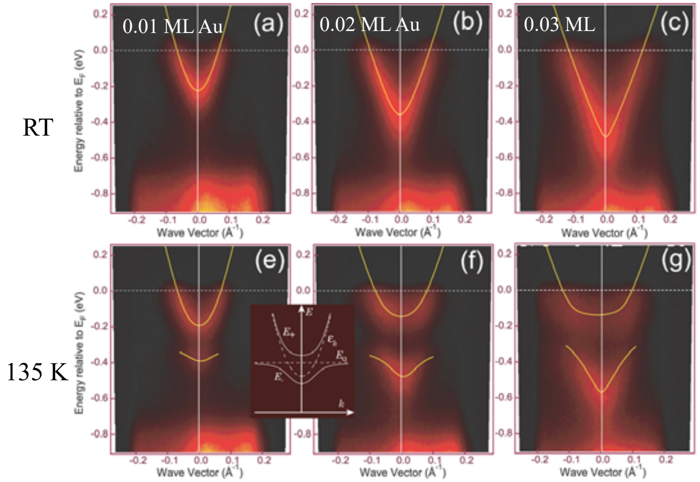
M. Aitani, Y. Sakamoto, T. Hirahara, M. Yamada, H. Miyazaki, M. Matsunami, S. Kimura, and S. Hasegawa:
Fermi level tuning of topological insulator thin films
Japanese Journal of Applied Physics **52**, 110112 (Oct, 2013)

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

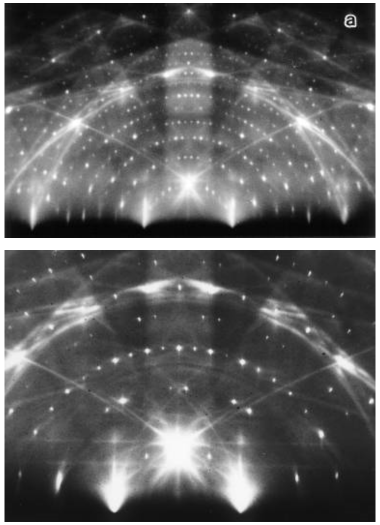
$$\boxed{\sigma = e \cdot \mu \cdot n}$$

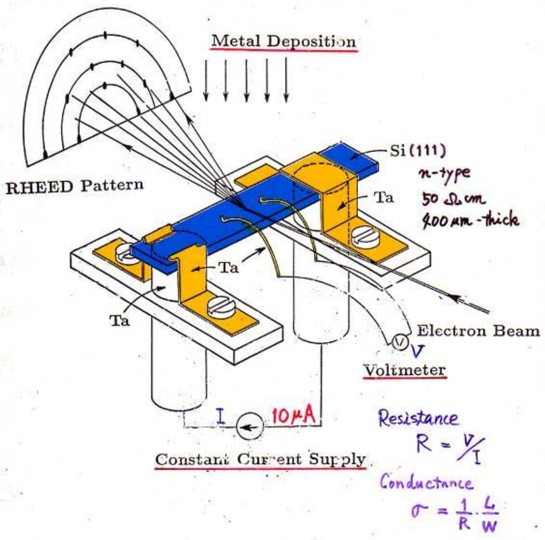




C. Liu, I. Matsuda, R. Hobara, and S. Hasegawa, Phys. Rev. Lett. **96**, 036803 (2006).

Macro-Four-Terminal Measurements in UHV

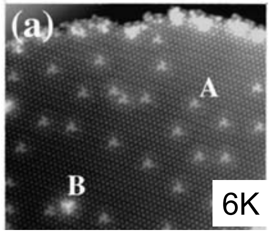




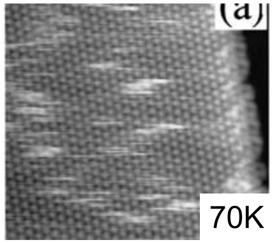
S. Hasegawa, et al., Phys. Rev. Lett. **68**, 1192 (1992)

Carrier Doping into Surface-State Band by Adatoms

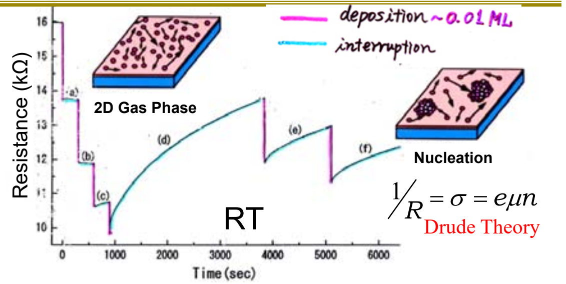
Y. Nakajima, *et al.*,
PRB **54**, 14 134(1996); **56**, 6782 (1997).



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

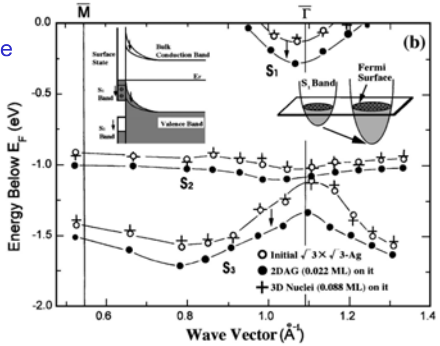


N. Sato, *et al.*, PRB **60**, 16 083(1999).



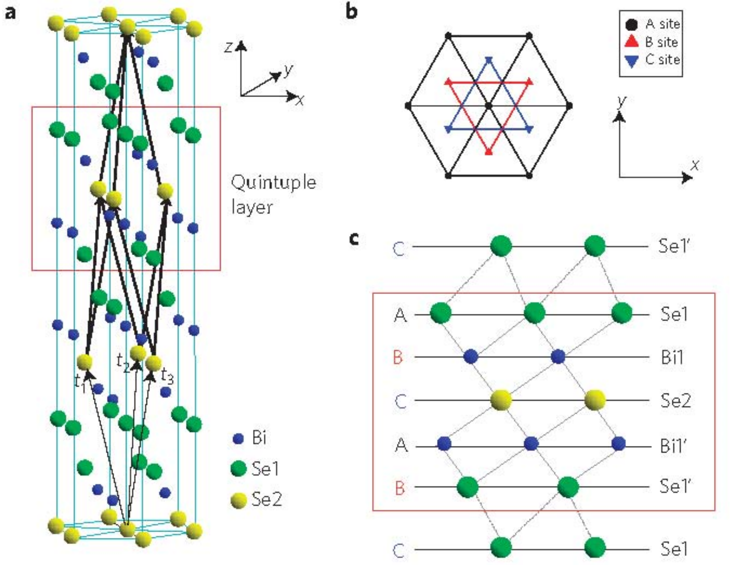
$$1/R = \sigma = e\mu n$$

Drude Theory



Crystal Structure of Bi_2Se_3 : Topological Insulator

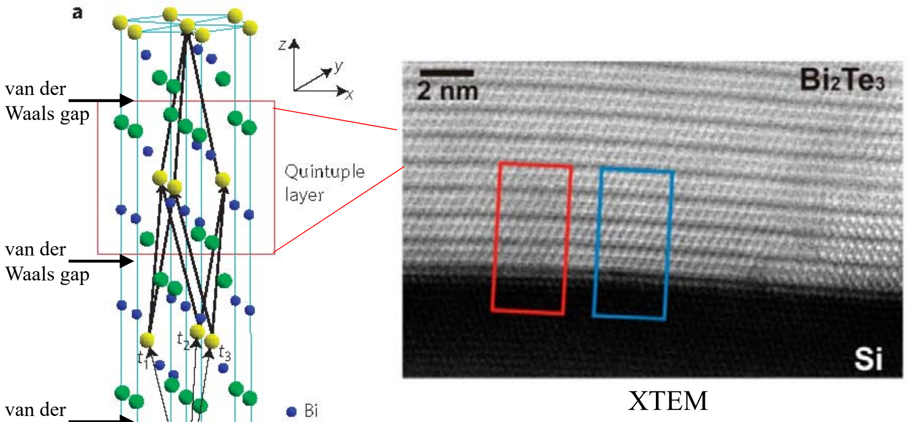
H. Zhang, et al., Nature Physics (May 2009)



Crystal Structure of Bi_2Se_3 (Bi_2Te_3)

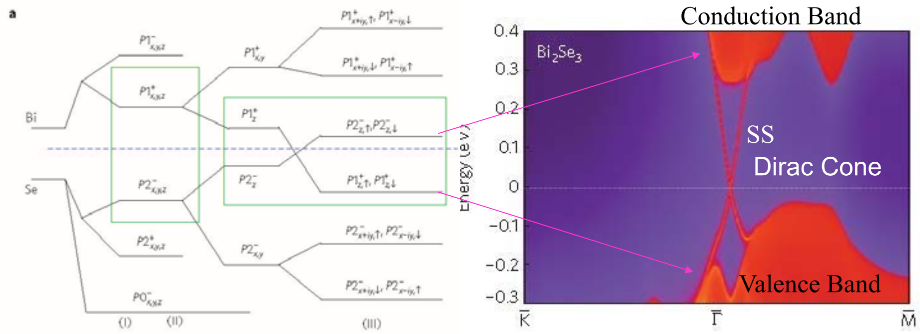
H. Zhang, *et al*., Nature Physics (2009)

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



Electronic States of Bi_2Se_3 (Theory)

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



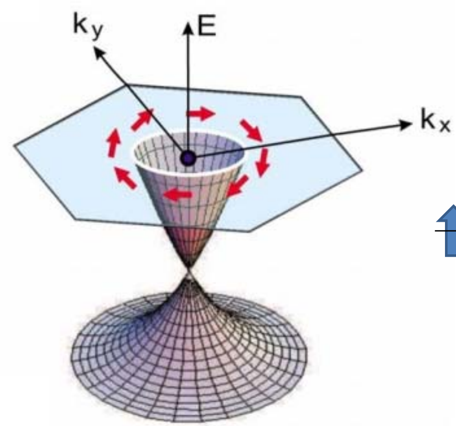
Isolated Atom
（Atomic Orbitals）

Atomic Bondings

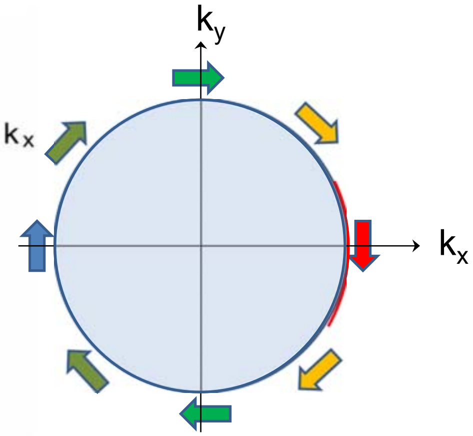
Split due to
Crystal filed

Spin-Orbit Intercation

Spin-Textured Fermi Surface＋Electric Field ⇒Current –Induced Spin Polarization



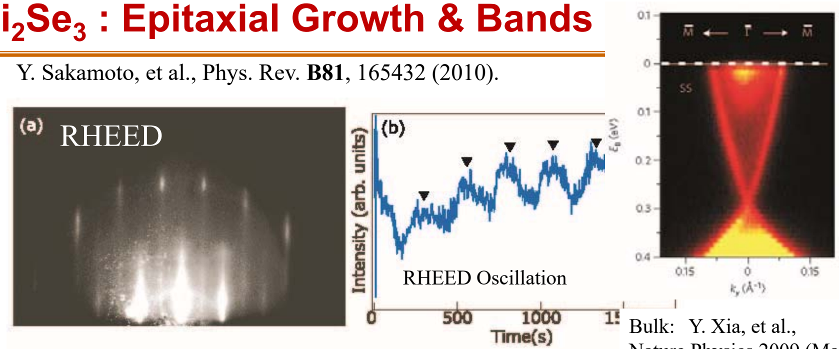
M. Z. Hasan and C. L. Kane,
Rev. Mod. Phys. **82**, 3045 (2010)



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

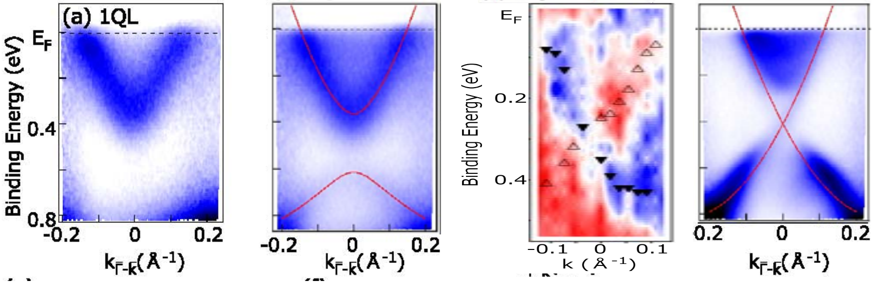
Bi_2Se_3 : Epitaxial Growth & Bands

Y. Sakamoto, et al., Phys. Rev. **B81**, 165432 (2010).

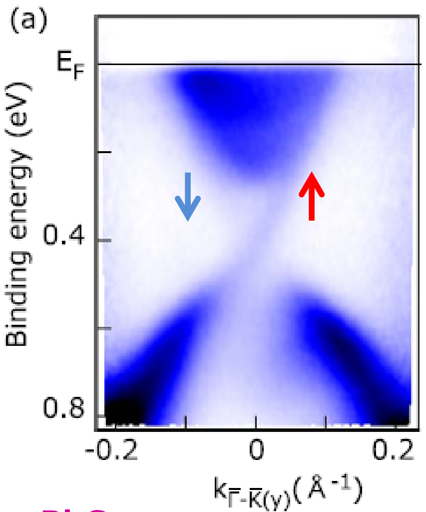


in
yer

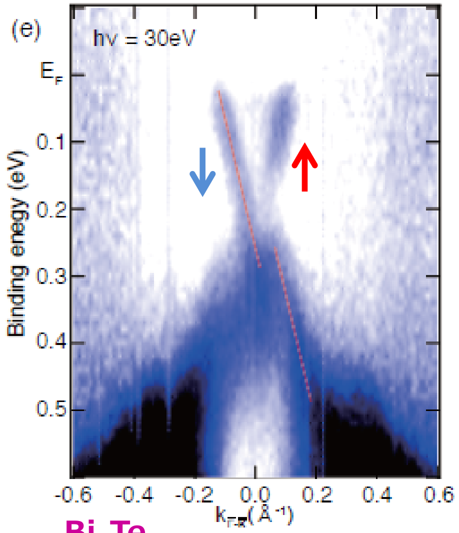
Bulk: Y. Xia, et al., Nature Physics 2009 (May)



Dirac Cones of Topological Insulators



Bi_2Se_3

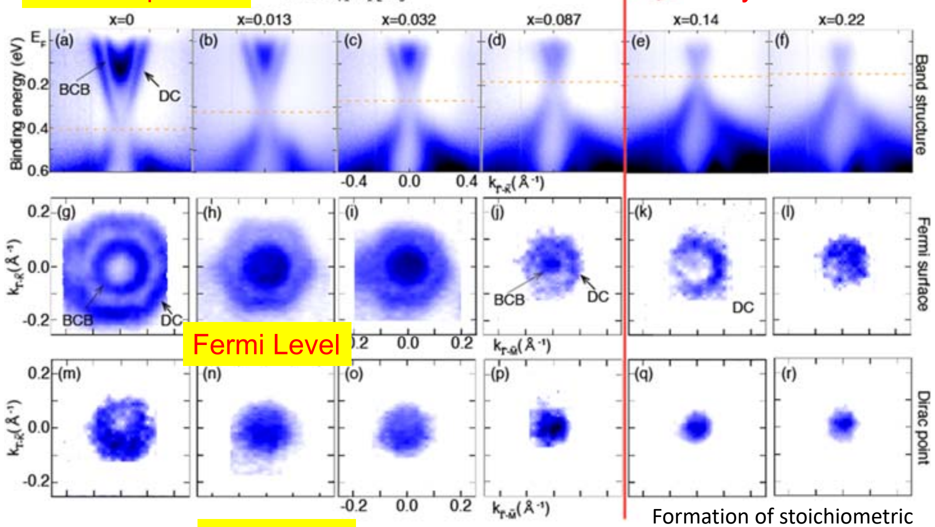


Bi_2Te_3

Hole-doping by Pb alloying in Bi_2Te_3

Band Dispersion

→ Truly bulk insulator



Fermi Level

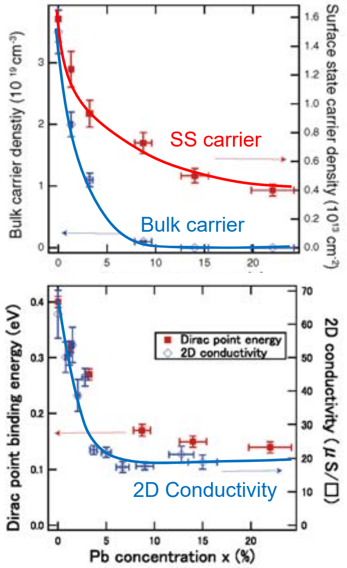
Dirac Point

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

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

Conductivity of a single Dirac-cone surface state

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



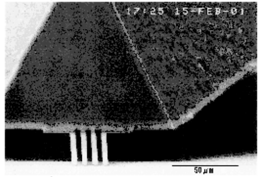
$$\boxed{\sigma_{SS} \approx 20\,\mu S \approx 0.5\frac{e^2}{h}}$$

Conductivity of
Single (Spin-split) Dirac Cone

Drude Model $\sigma = e\mu n$

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

$\Rightarrow$ Mobility $\boxed{\mu = 30 \text{ cm}^2/\text{Vs}}$



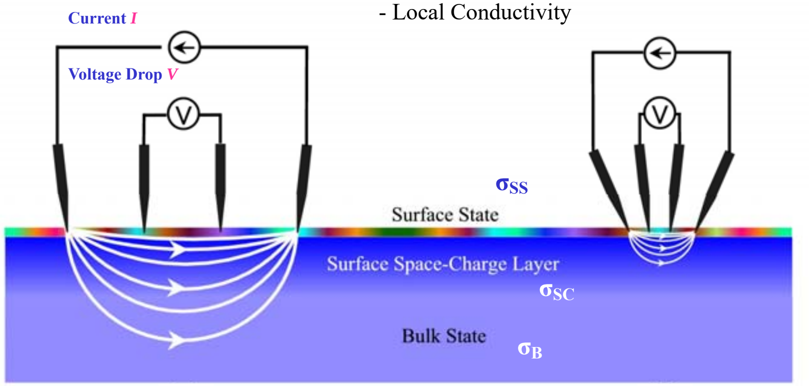
Not high mobility!

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 $\sigma_{meas} = \sigma_{SS} + \sigma_{SC} + \sigma_{B}$
- Local Conductivity



Macro-4-Point Probe

Bulk-Sensitive
(Surface -Insensitive)

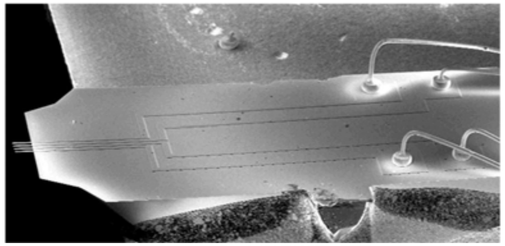
Micro-4-Point Probe

Surface -Sensitive

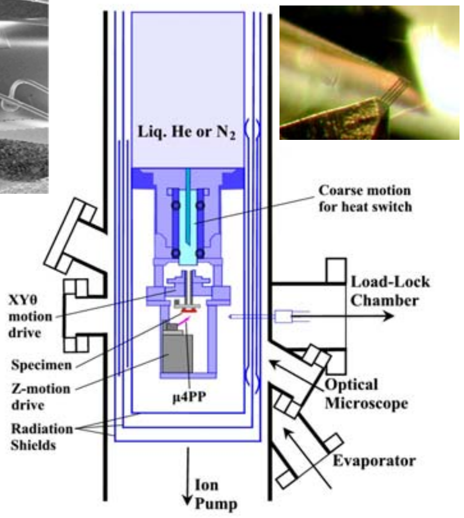
Temperature-Variable Monolithic Micro-Four-Point Probe

I. Shiraki, et al., Surf. Rev. Lett. **7** (2000) 533.
C. L. Petersen, 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.

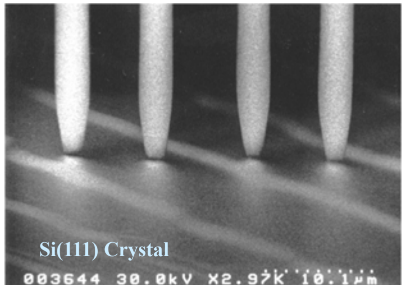
Developed at Denmark Technical University



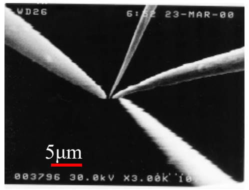
Commercially available;
http://www.capres.com

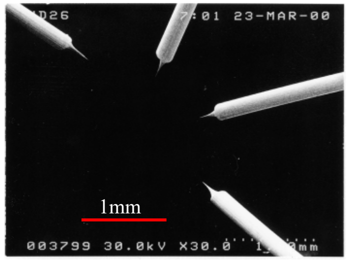


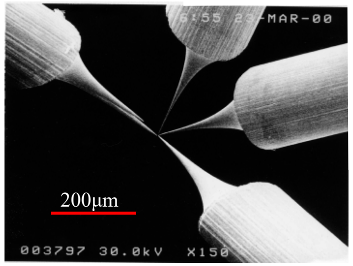
Contacting to Si(111) sample surface in SEM



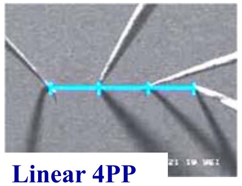
Conductivity Measurements by 4-Tip STM



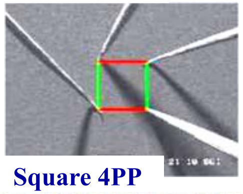




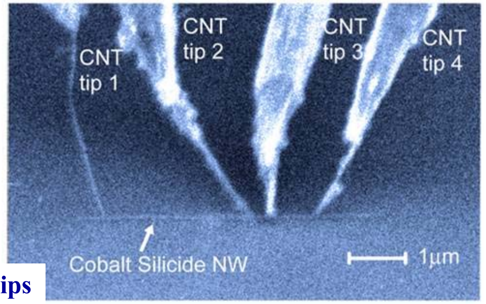
W-Tips



Linear 4PP



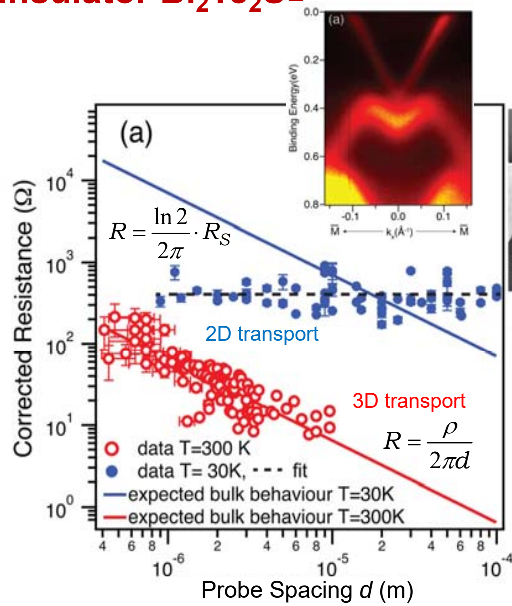
Square 4PP

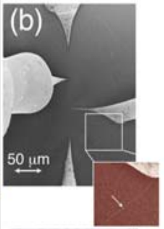


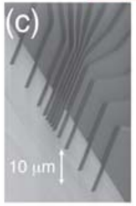
CNT Tips

Surface-Dominated Transport on a Bulk Topological Insulator Bi_2Te_2Se

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







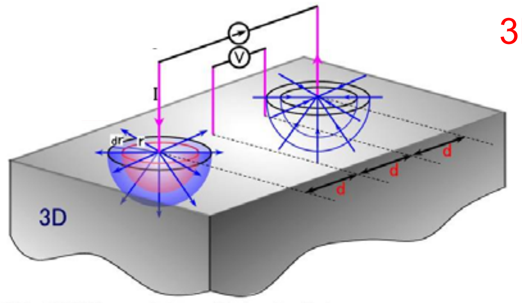
LT: Bulk carriers are frozen out.
⇒ SS transport only

⇒ Mobility

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

Fairly high mobility!

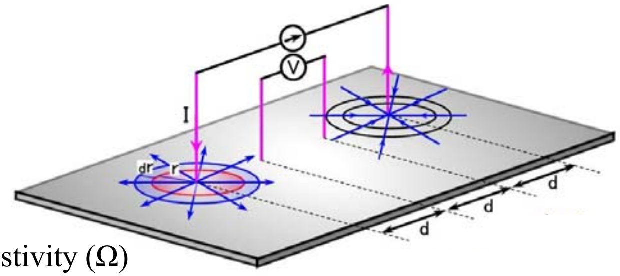
Current Distribution vs. Probe-Spacing Dependence of Resistance



3D transport

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

ρ : 3D Resistivity (Ωcm

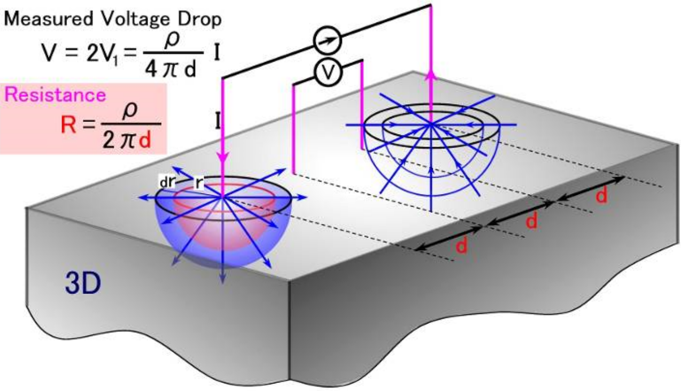


2D transport

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

R_S: Sheet Resistivity (Ω)

Four-Point Probe Resistance for a 3D sample



Measured Voltage Drop

$$V = 2V_1 = \frac{\rho}{4\pi d}\,I$$

Resistance

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

Potential Difference between the two hemispheres

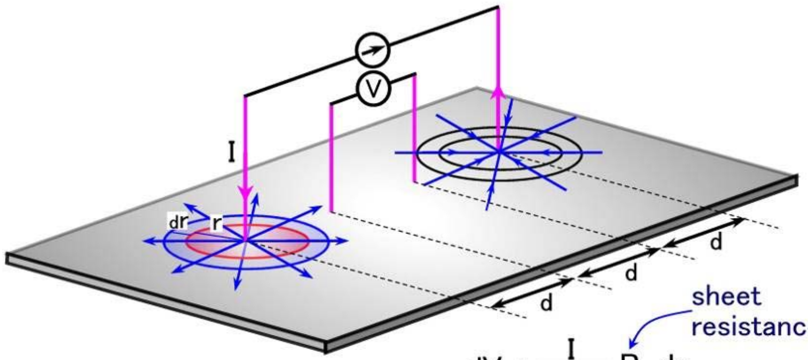
$$dV_1 = \frac{I}{2\pi r^2}\,\rho\,dr$$

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]

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

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

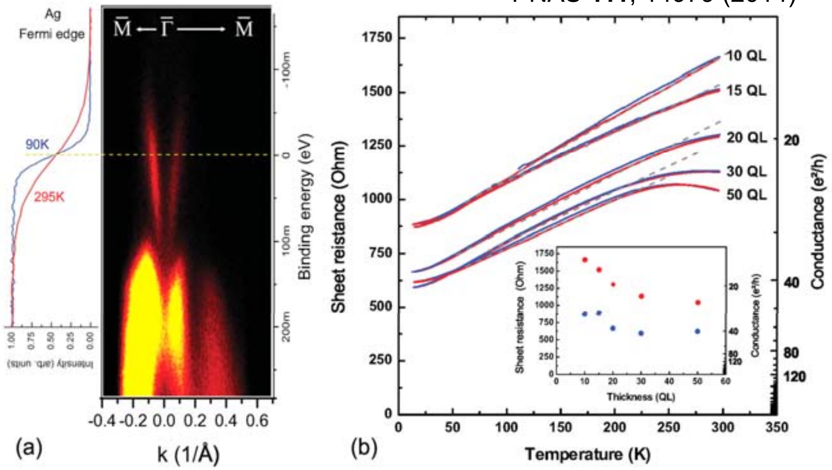
$$= \frac{R_S I}{2\pi} \cdot \ln \frac{2\cancel{d}}{\cancel{d}} = \frac{\ln 2}{2\pi}\,R_S I$$

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

$R = \frac{\ln 2}{2\pi} R_S$: 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 $\boxed{\mu = 4{,}600\ \mathrm{cm^2/Vs}}$ Very high mobility!

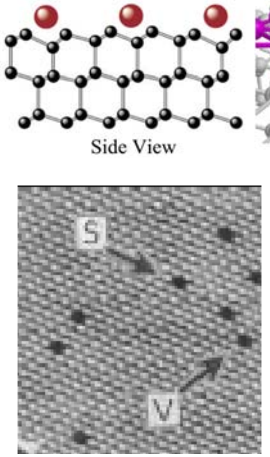
Surface-State Superconductivity

M. Yamada, T. Hirahara, and S. Hasegawa:
Magnetotransport measurements of
a superconducting surface state of In- and
Pb-induced structures on Si(111)
Phys. Rev. Lett. **110**, 237001 (Jun, 2013).

Indium-adsorbed Silicon (111) Surface

Insulator

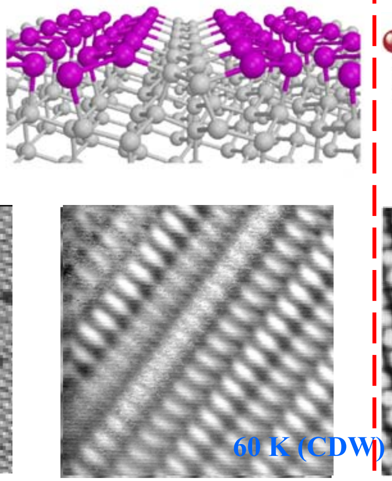
$\sqrt{3}\times\sqrt{3}$



A. A. Sarranin, et. al.

Quasi-1D Metal

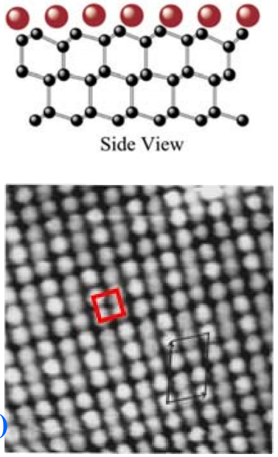
4×1



H. Y. Yeom, et al., PRL **82**, 4898 (1999)

2D Metal

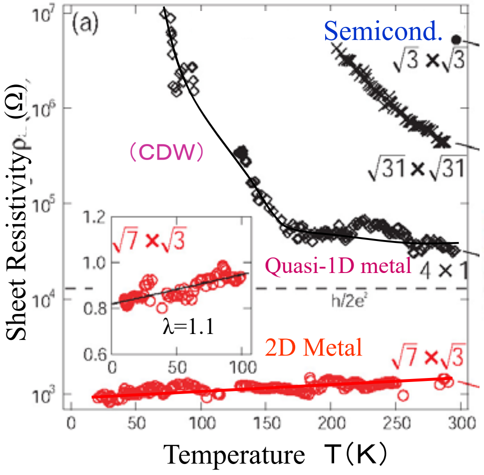
$\sqrt{7}\times\sqrt{3}$

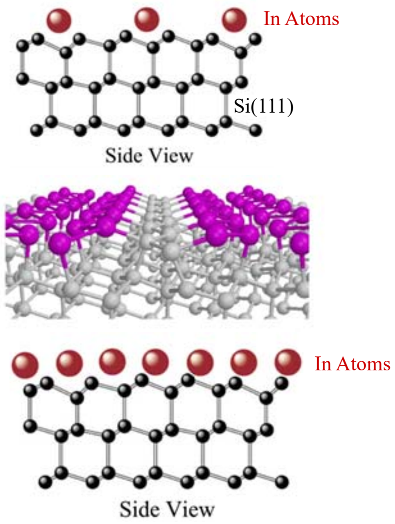


S. L. Surnev, et. al.

Electrical Resistance of (Sub)Monolayer In-adsorbed Si

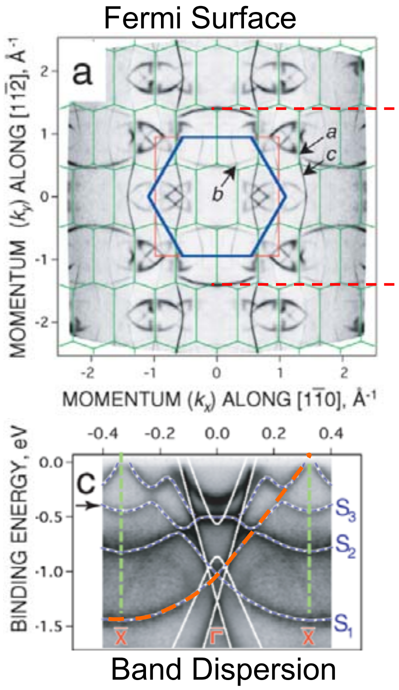
S. Yamazaki, et. al., Phys. Rev. Lett., **106**, 116802 (2011).





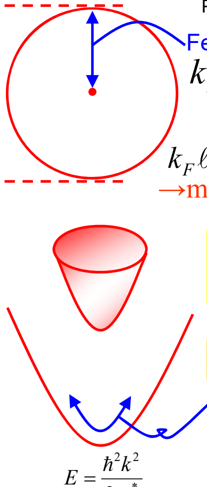
√7×√3: Suggestions of Superconductivity

- **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)



Fermi wave number

$k_F = 14\mathrm{nm}^{-1}$(very large)

Circular Fermi surface

$k_F \ell \cong 20 >> 1$

→metallic conduction

Isotropic two dimensional nearly-free electron gas

Parabolic dispersion relation

Effective mass

$m^* = 1.1 m_e$

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

$$D_{2D} = \frac{m^*}{\pi\hbar^2} = 4.6[\mathrm{eV}^{-1}\mathrm{nm}^{-2}]$$

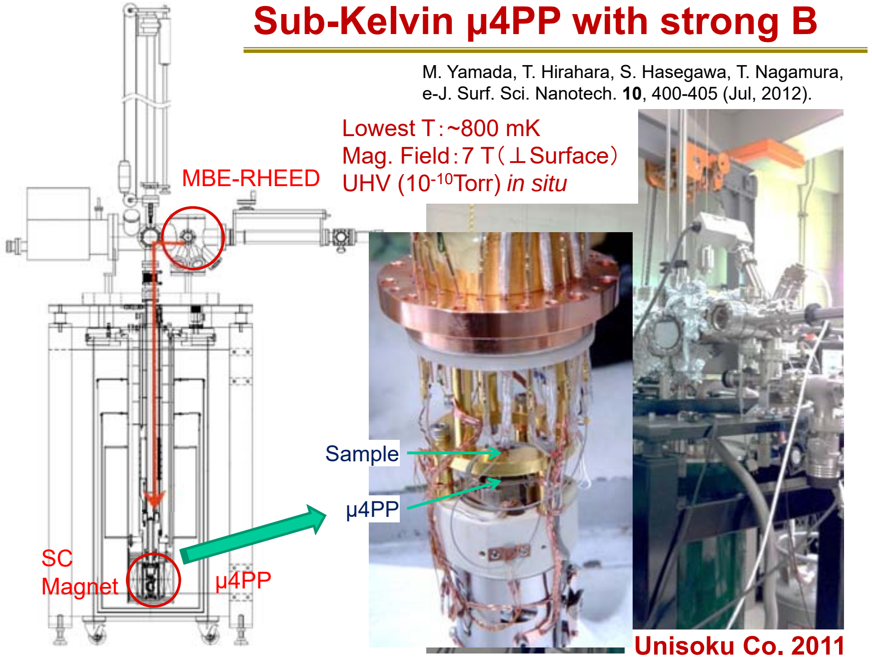
$$n = 3.1\times10^{15} cm^{-2}$$

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

Sub-Kelvin μ4PP with strong B

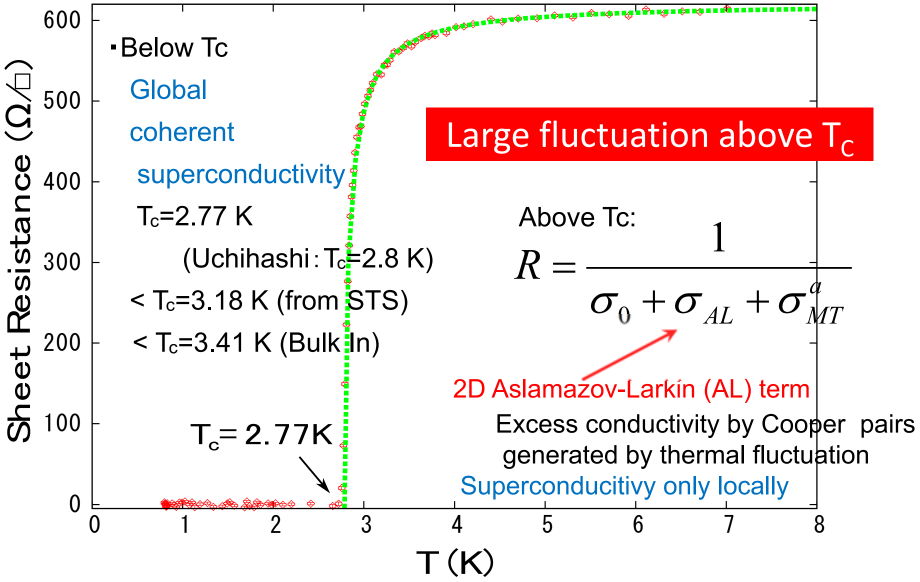
M. Yamada, T. Hirahara, S. Hasegawa, T. Nagamura,
e-J. Surf. Sci. Nanotech. **10**, 400-405 (Jul, 2012).

Lowest T : ~800 mK
Mag. Field : 7 T (⊥ Surface)
UHV (10^{-10}Torr) *in situ*



Unisoku Co. 2011

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



- Below Tc

Global coherent superconductivity
T_c=2.77 K
(Uchihashi : T_c=2.8 K)
< T_c=3.18 K (from STS)
< T_c=3.41 K (Bulk In)

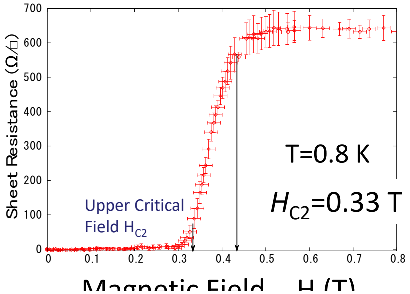
Large fluctuation above T_C

Above Tc:

$$R = \frac{1}{\sigma_0 + \sigma_{AL} + \sigma^{a}_{MT}}$$

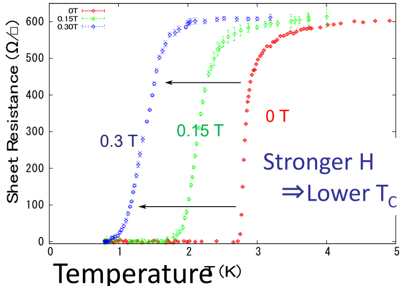
2D Aslamazov-Larkin (AL) term
Excess conductivity by Cooper pairs generated by thermal fluctuation
Superconducitivy only locally

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

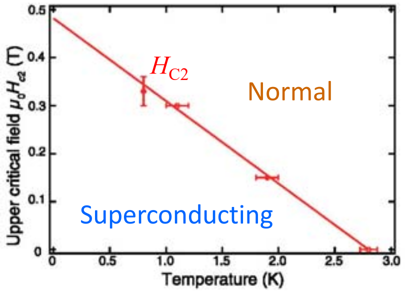


T=0.8 K

H_{C2}=0.33 T



Stronger H
⇒Lower T_C



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

Very high H_c (=short ξ)

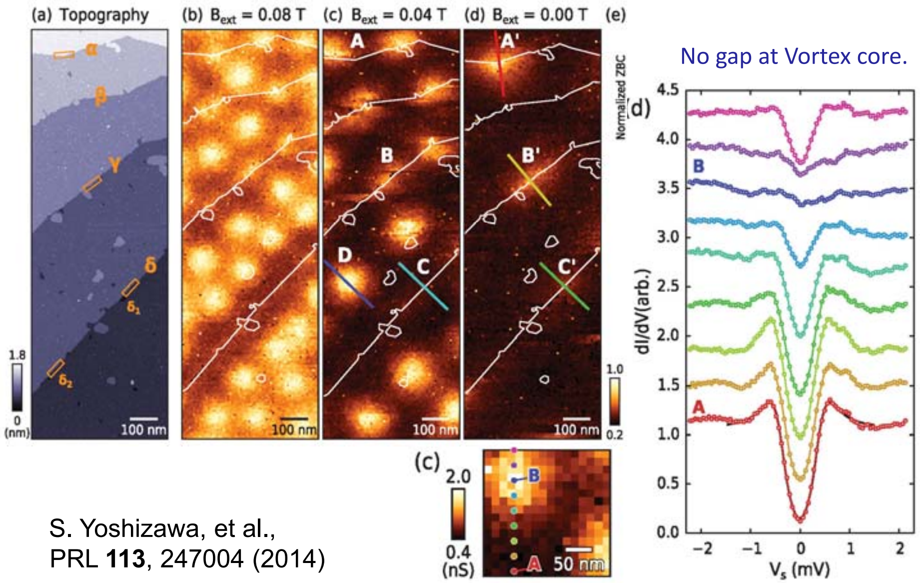
Strong Critical Field $H_{c2}(0)$~5000 Oe

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



No gap at Vortex core.

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

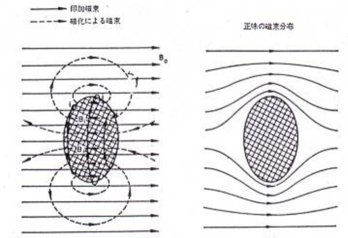
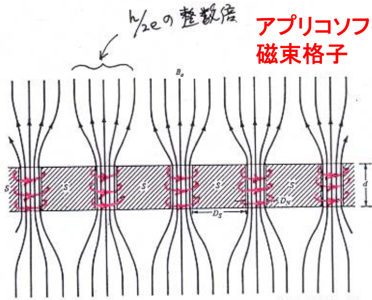


図 2.1　完全反磁性体の近傍の磁束の分布

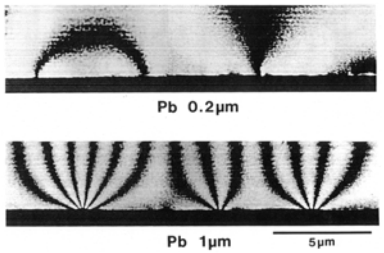
マイスナー効果

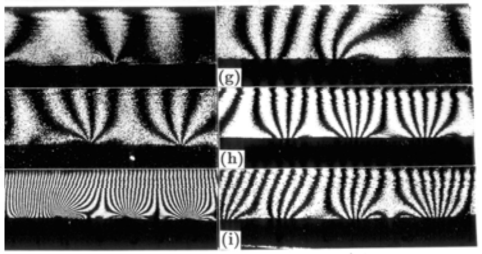


3-6 図　中間状態で，磁束が層状の常伝導部分を貫いている様子の模式図．遠く離れた所では磁束密度は B_a であり，平板の断面内ではゼロかあるいは $h_n(\approx H_c)$ である．

アプリコソフの
磁束格子

S. Hasegawa, et al., Phys. Rev. B **43**, 7631(1991)

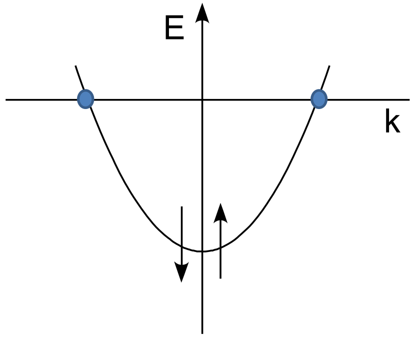




d = 1.0 μm　　d = 1.7 μm

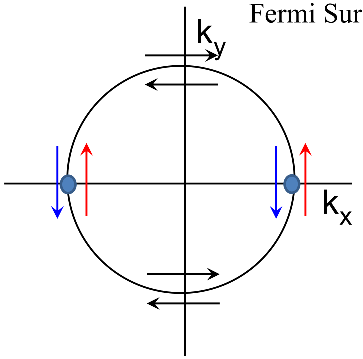
Cooper Pairs in Free-Electron Band

Band Dispersion



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

Fermi Surface

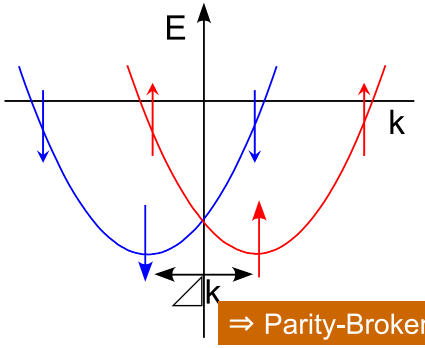


Singlet　$|k\uparrow\rangle|-k\downarrow\rangle-|k\downarrow\rangle|-k\uparrow\rangle$

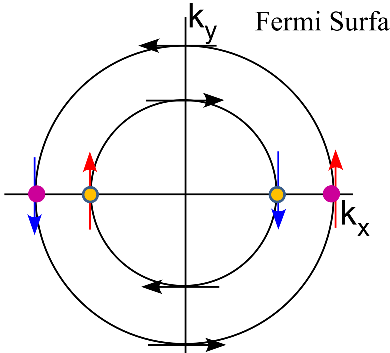
Triplet　$|k\uparrow\rangle|-k\uparrow\rangle,\ |k\downarrow\rangle|-k\downarrow\rangle,$
$|k\uparrow\rangle|-k\downarrow\rangle+|k\downarrow\rangle|-k\uparrow\rangle$

Spin Split and Cooper Pairs in Free-Electron Band

Band Dispersion



Fermi Surface



⇒ Parity-Broken Superconductors

$\hbar^2$

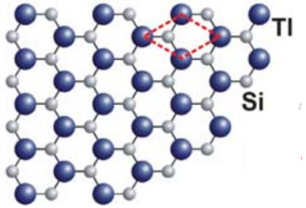
●Outer　$|k\uparrow\rangle|-k\downarrow\rangle$

$$\underbrace{|k\uparrow\rangle|-k\downarrow\rangle}=\frac{1}{2}\underbrace{\Big(\underbrace{|k\uparrow\rangle|-k\downarrow\rangle}-\underbrace{|k\downarrow\rangle|-k\uparrow\rangle}\Big)}_{\text{Singlet (s-wave)}}+\frac{1}{2}\underbrace{\Big(\underbrace{|k\uparrow\rangle|-k\downarrow\rangle}+\underbrace{|k\downarrow\rangle|-k\uparrow\rangle}\Big)}_{\text{Triplet(p-wave)}}$$

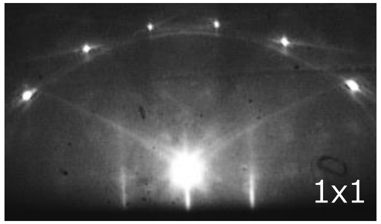
Sample: Si(111)–√3x√3-(Tl, Pb) Surface Superstructure

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

Si(111)−1x1-Tl



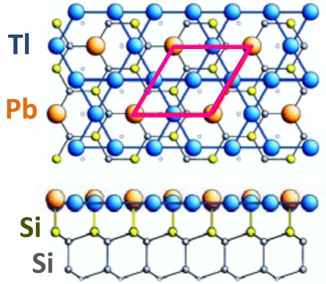
Tl 1ML on 7x7 @ 600K

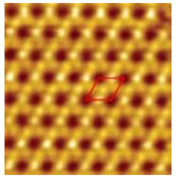


⇩

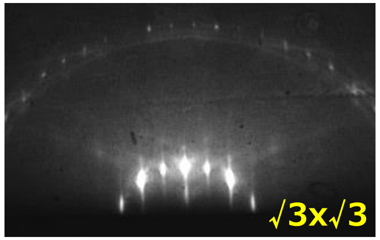
+Pb 1/3 ML @ 300K

Si(111)−√3x√3-(Tl, Pb)





⟵ Mono-Layer

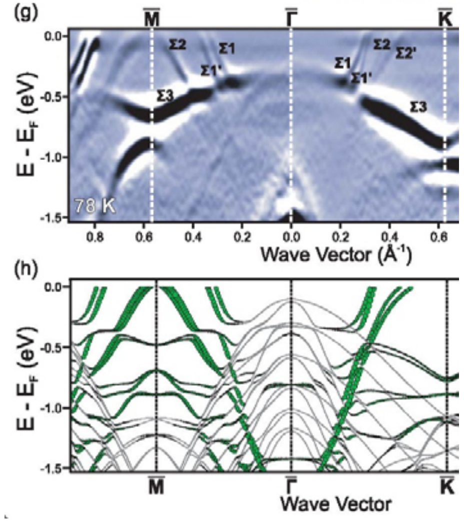


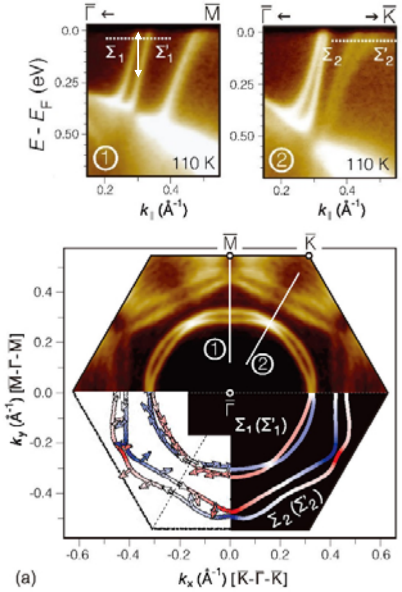
Si(111)-√3 × √3-(Tl, Pb) :Rashba-type SS

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

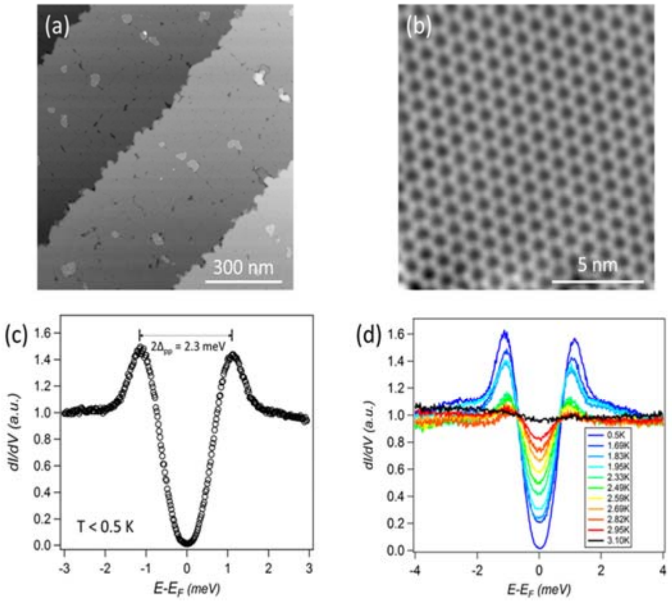
ARPES+Theory

⇒ Spin-Split Surface-State Bands





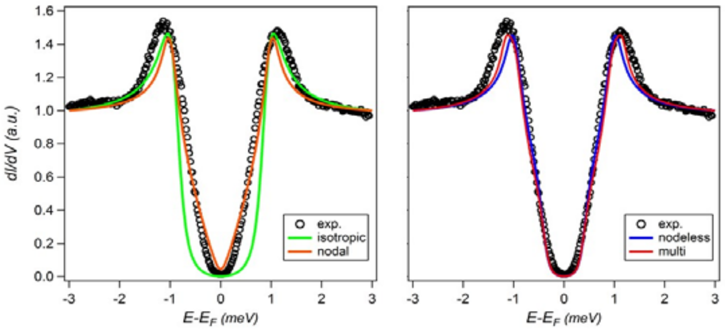
STS below T_C on Si(111)-√3 × √3-(Tl,Pb) w/o B



Fitting STS Spectra by Theory

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

$$\frac{\mathrm{d}I_{\mathrm{ns}}}{\mathrm{d}V} = \rho_{\mathrm{t}}(0)\rho_{\mathrm{n}}(0)\int_{-\infty}^{\infty} \mathrm{Re}\left\{\frac{|E - i\Gamma|}{\sqrt{((E - i\Gamma)^2 - \Delta^2)}}\right\}\left[\frac{\exp[(E+eV)/k_{\mathrm{B}}T]}{k_{\mathrm{B}}T\{1+\exp[(E+eV)/k_{\mathrm{B}}T]\}^2}\right]\mathrm{d}E$$



s-wave (isotropic gap)

$\Delta = const.$ (1.0 meV)

anisotropic gap

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

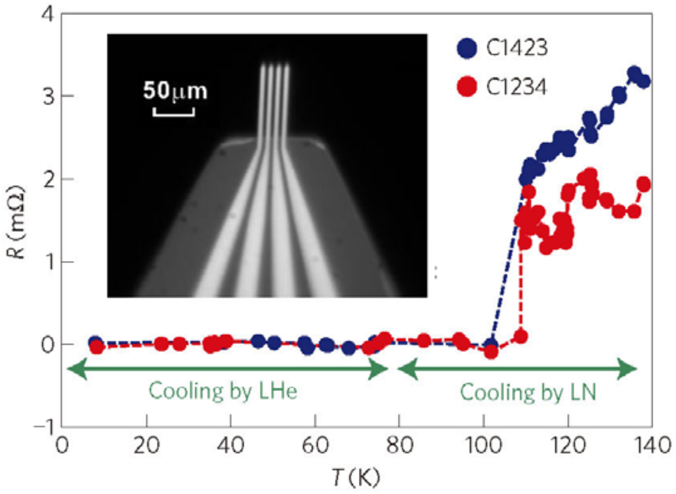
$\Delta_0 = 0.47$ meV

$\Delta_1 = 0.84$ meV

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

J-F. Ge, *et al.*,
Nat. Mat. (Nov, 2014)

measured in situ by μ4PP in UHV



Cf: Bulk FeSe

T_C = 9.4 K

Spin Transport at Surfaces

T. Tono, T. Hirahara, and S. Hasegawa:
In situ transport measurements on ultrathin Bi(111) films using a magnetic tip: Possible detection of current-induced spin polarization in the surface states
New J. Phys. **15**, 105018 (Oct 2013)

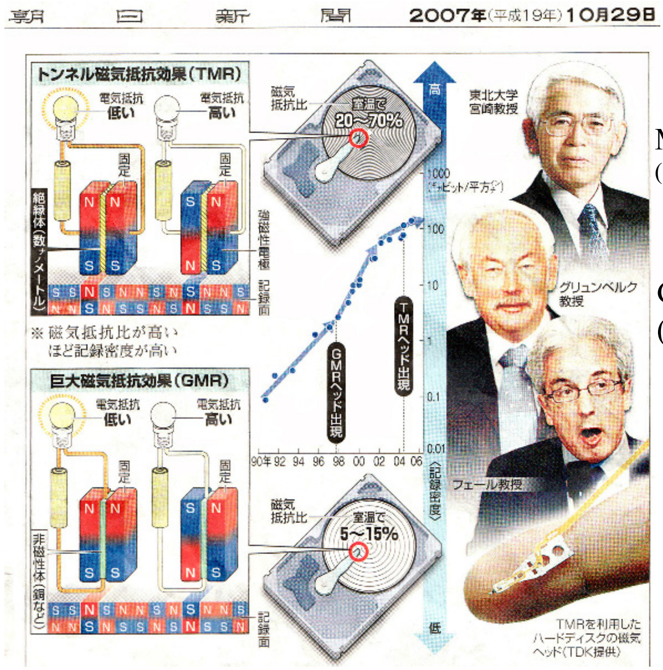
Utilizing Spins

- 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

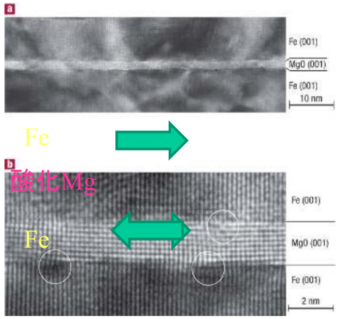
Discovery of Giant MagnetoResistance (GMR) Effect
巨大磁気抵抗効果の発見 ⟶ 磁気ヘッド（ハードディスクの小型化・高密度化）
Magnetic Head (HDs becomes smaller and high-density)

Albert Fert
France
南パリ大学

b. 1938

Peter Grünberg
Germany
Julich Inst

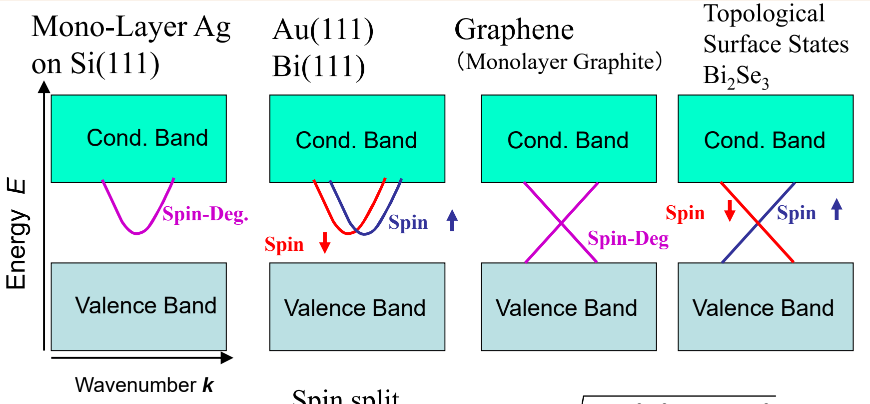
b. 1939



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

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

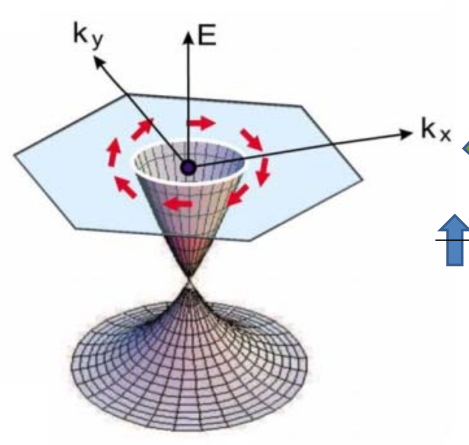
$$\Big\Downarrow \quad m = 0$$

Relativistic

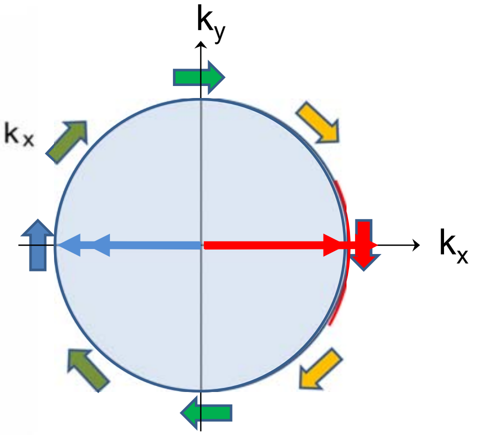
$$E = \pm pc = \pm c\hbar k$$

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)

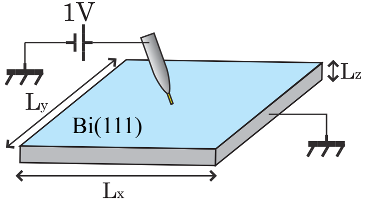


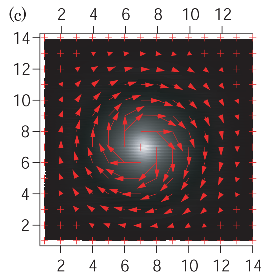
Spin-Textured Fermi Surface
フェルミ面のスピン繊維構造

Current-induced Spin Polarization

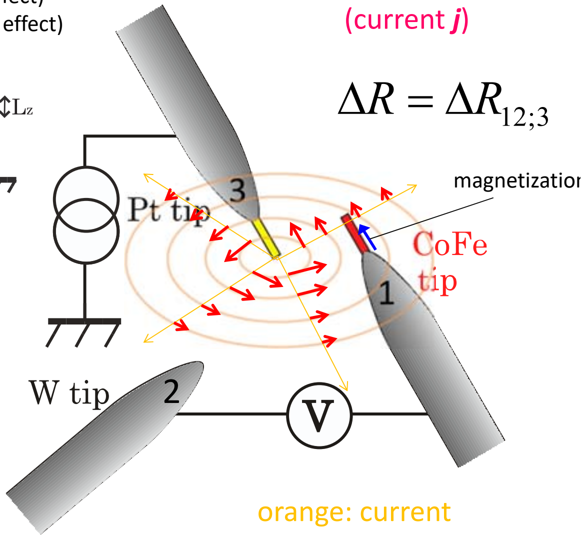
- No macroscopic or local magnetic field effect (Hall effect)
- No edge (spin Hall effect)

Spin ***s*** ⊥ wavevector ***k***
(current ***j***)



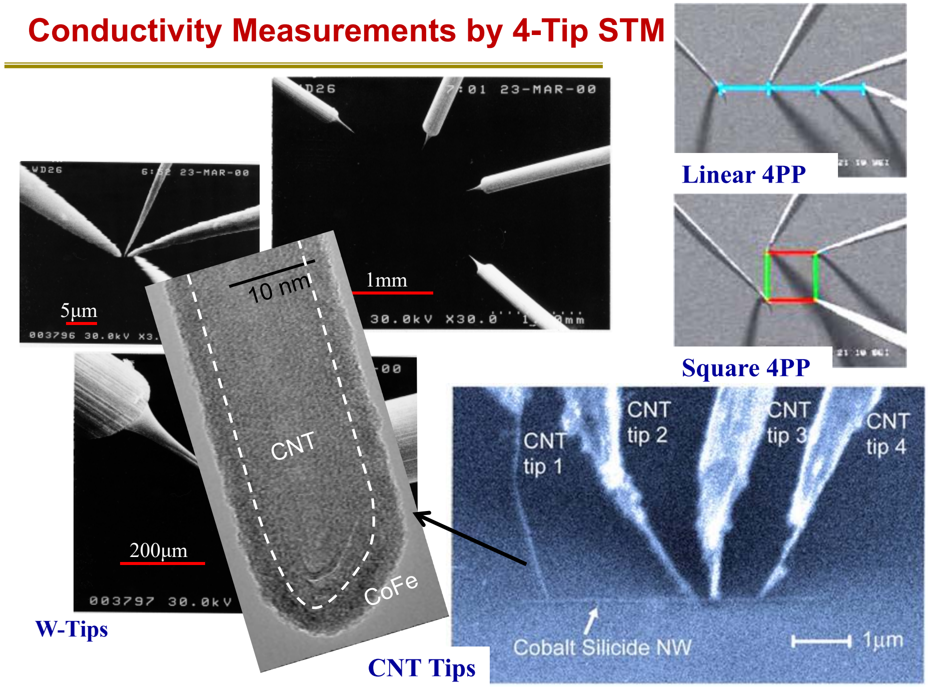


$$\Delta R = \Delta R_{12;3}$$



orange: current
red: local spin polarizatior

Conductivity Measurements by 4-Tip STM



Signal of Spin Orientation on Bi(111) Surface

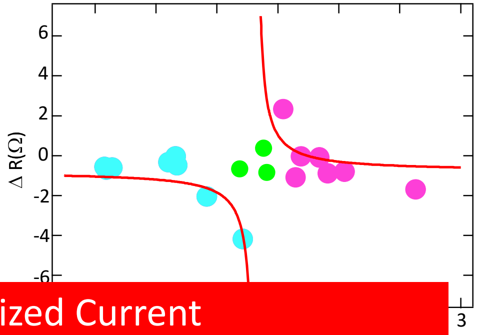
$$\vec{s} = -\frac{\hbar}{2}\tanh\left(\frac{\alpha_R \delta k}{k_B T}\right)\hat{\theta}$$

$$\delta\vec{k} = \frac{e\tau}{\hbar}\vec{E}$$

$$\boxed{\vec{s} \approx -\frac{\hbar}{2}\frac{\alpha_R}{k_B T}\frac{e\tau}{\hbar}E\hat{\theta}}$$

$$\vec{E} = \frac{I}{}\cdot\frac{1}{}\hat{d}$$

M. Liu *et al.*,

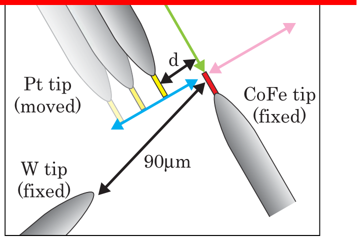


Detecting Spin-Polarized Current on a Non-magnetic Surface ?

$$\Delta R = \eta\frac{}{AN_0}\cdot\frac{}{k_B T}\cdot\frac{}{\hbar}\cdot\frac{}{2\pi\sigma}\cdot\frac{}{d}\approx\eta\cdot\frac{}{2eI}$$

$$\text{Spin Density } s_D \approx N_0 e\frac{\mu_\uparrow - \mu_\downarrow}{2}$$

d	10 nm	100 nm	1 μm
$\mu_\uparrow - \mu_\downarrow$	300 μV	30 μV	3 μV
s_D	800 μm⁻²	80 μm⁻²	8 μm⁻²



Spin Hall Effect

磁場の印加が不要

No external magnetic field

非磁性物質（SOCが強い）

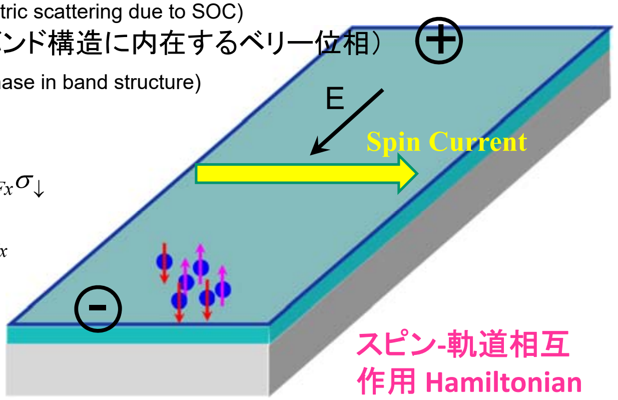
Non-magnetic materials(strong SOC)

- ・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

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

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

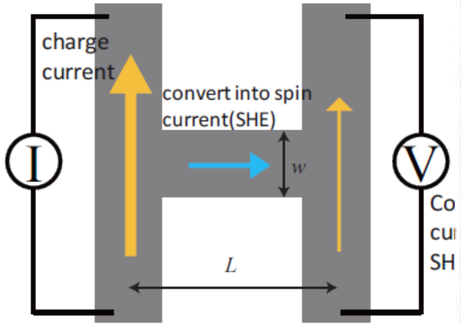


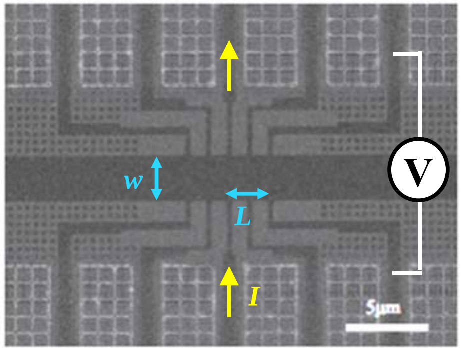
スピン-軌道相互作用 Hamiltonian

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

Detecting Spin Hall Effect

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

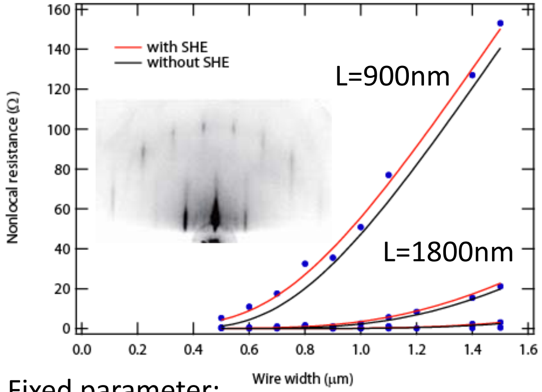


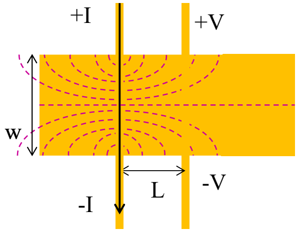


Current $\xrightarrow{\text{SHE}}$ Spin Flow $\xrightarrow{\text{ISHE}}$ Current/ Voltage → Electrical Detection

It should be *L* < *Ls* (Spin relaxation length).

Results on Bi_2Se_3 thin film (8QL) at RT





Fixed parameter:

$$\rho_{2D} = 805\Omega$$

fitting parameter

Spin-Hall Angle

$$\gamma = \sigma_s/\sigma_c = 0.032 \pm 0.005$$

Spin Diffusion Length

$$l_s = 0.23 \pm 0.09\,\mu m$$

From Ohm's law

$$R_c = \frac{2\rho_{2D}}{\pi}\log\coth\left(\frac{\pi L}{2w}\right)$$

From SHE

$$R_{nl}(x) = \frac{1}{\pi}\left(\frac{\beta_s}{\sigma}\right)^2\frac{1}{\sigma}\int_0^\infty \cos\left(\frac{x}{l_s}k\right)\left(1-\exp\left(-\frac{w}{l_s}\sqrt{k^2+1}\right)\right)\tanh\left(\frac{k}{2}\frac{w}{l_s}\right)\frac{kdk}{k^2+1}$$