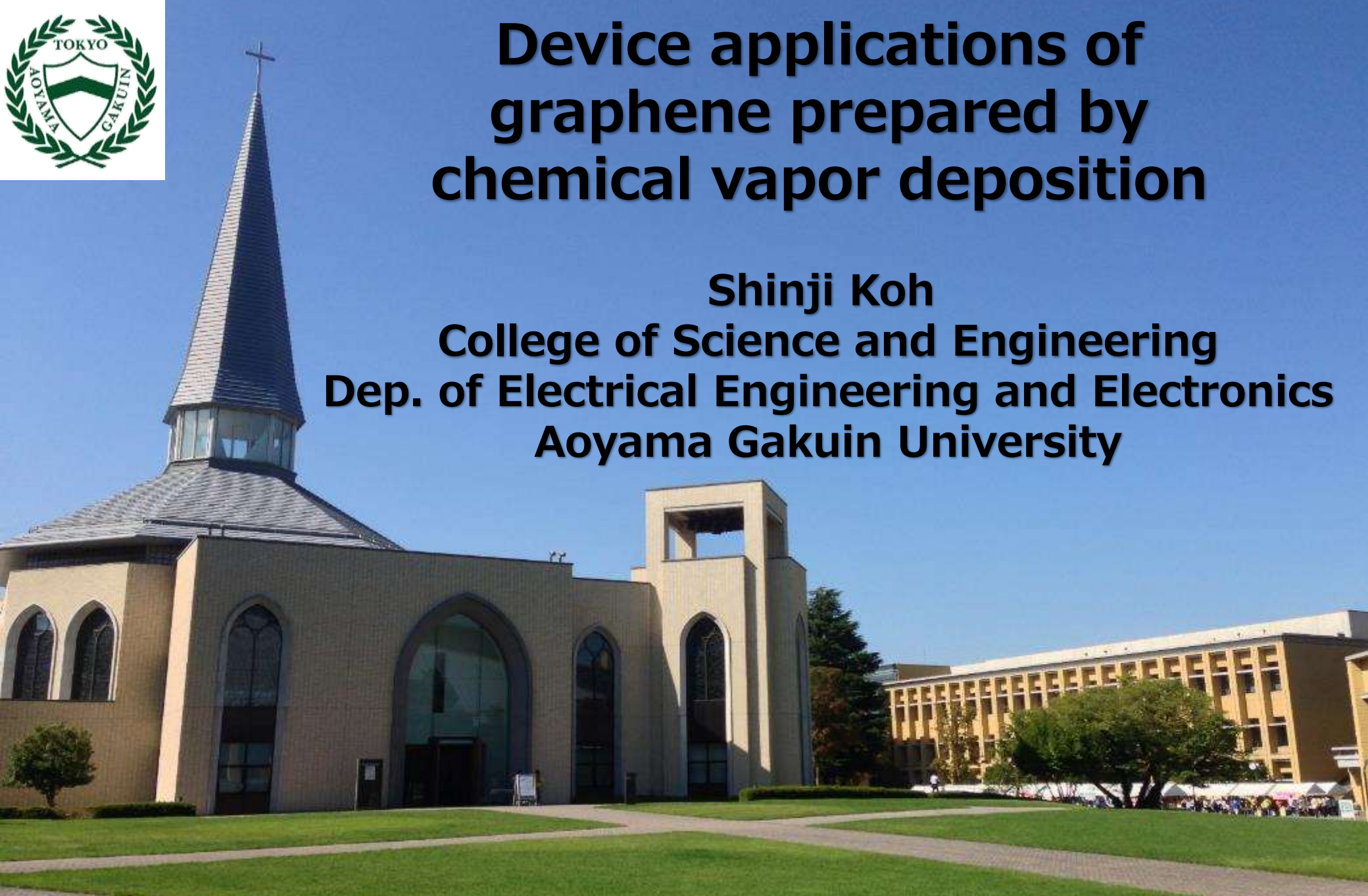




Device applications of graphene prepared by chemical vapor deposition

Shinji Koh

**College of Science and Engineering
Dep. of Electrical Engineering and Electronics
Aoyama Gakuin University**



Aoyama Gakuin University

Private University

Founded in **1874**

Faculty member	~600
Undergraduates	~17,000
Graduate students	~1,200

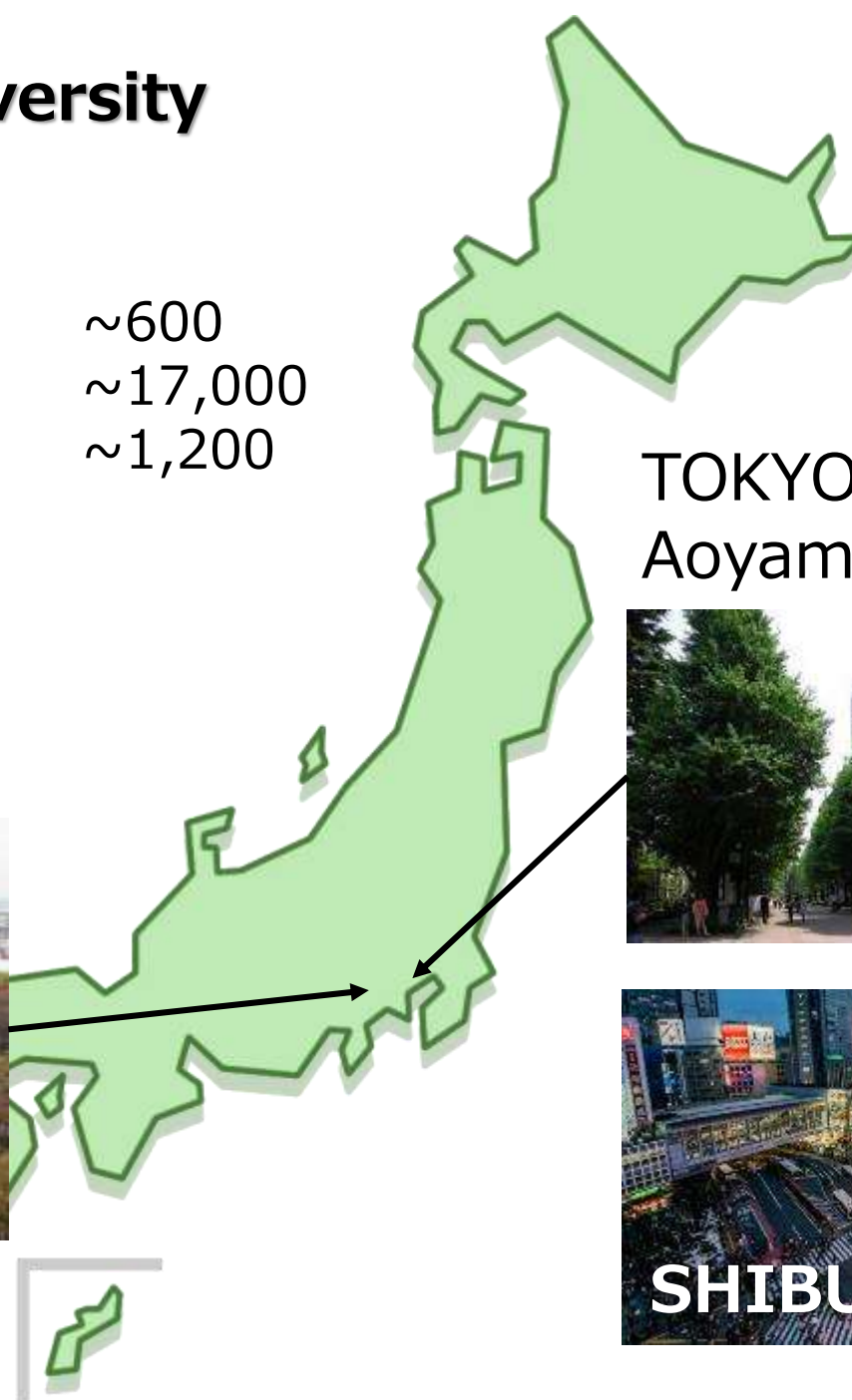


TOKYO
Aoyama campus

KANAGAWA
Sagamihara campus
I work here!

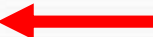


SHIBUYA



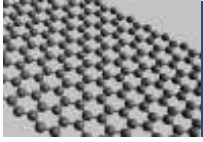
Top academic institutions by normalized WFC

The tables show Japan's leading institutions ranked by normalized weighted fractional count (WFC), an in proportion of total output in the natural sciences (NS) from 2012 to 2017. This is derived by dividing an ins total number of natural sciences articles in Scopus (Scopus NS articles 2012–2017). Also listed are an ins 2012–2017) and the percentage of its total articles in Scopus that are in the natural sciences (NS articles

Rank	Institution	Normalized WFC 2012–2017
1	Gakushuin University	0.0938
2	The University of Tokyo (UTokyo)	0.0680
3	Konan University	0.0611
4	Kyoto University	0.0577
5	Aoyama Gakuin University 	0.0575
6	Osaka University	0.0574
7	Nara Institute of Science and Technology (NAIST)	0.0563
8	Okinawa Institute of Science and Technology Graduate University (OIST)	0.0561
9	Tokyo Institute of Technology (Tokyo Tech)	0.0547
10	Nagoya Institute of Technology (NITech)	0.0491
11	Nagoya University	0.0478

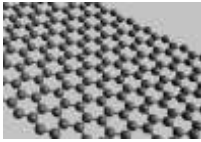
Nature Index 2018

5th position Japanese academic institution



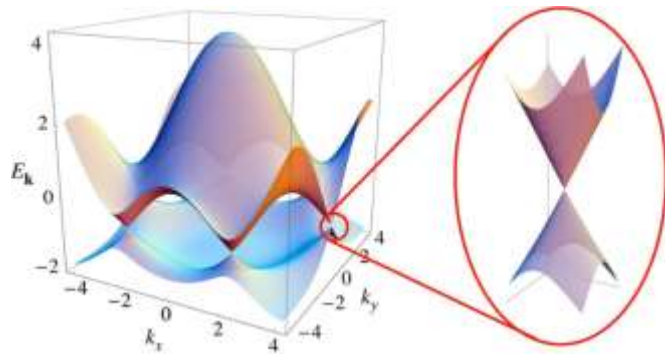
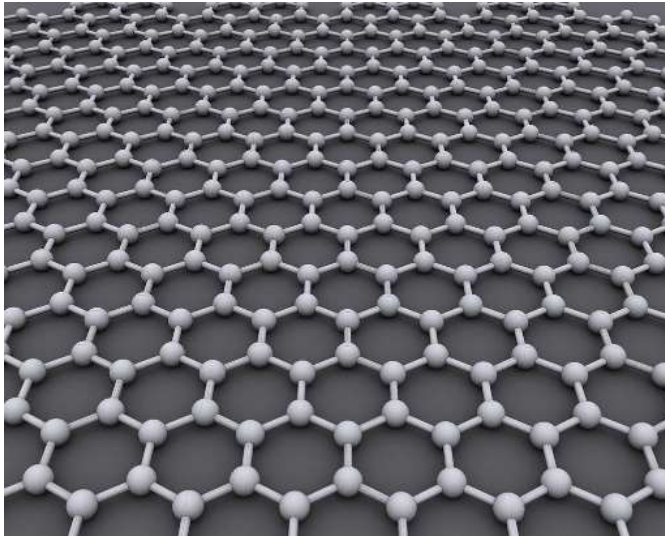
OUTLINE

- 1. CVD growth of graphene on Ir(111)/sapphire**
- 2. Device applications of CVD graphene grown on Cu**
 - a. Optically transparent antennas**
 - b. Free chlorine sensors**



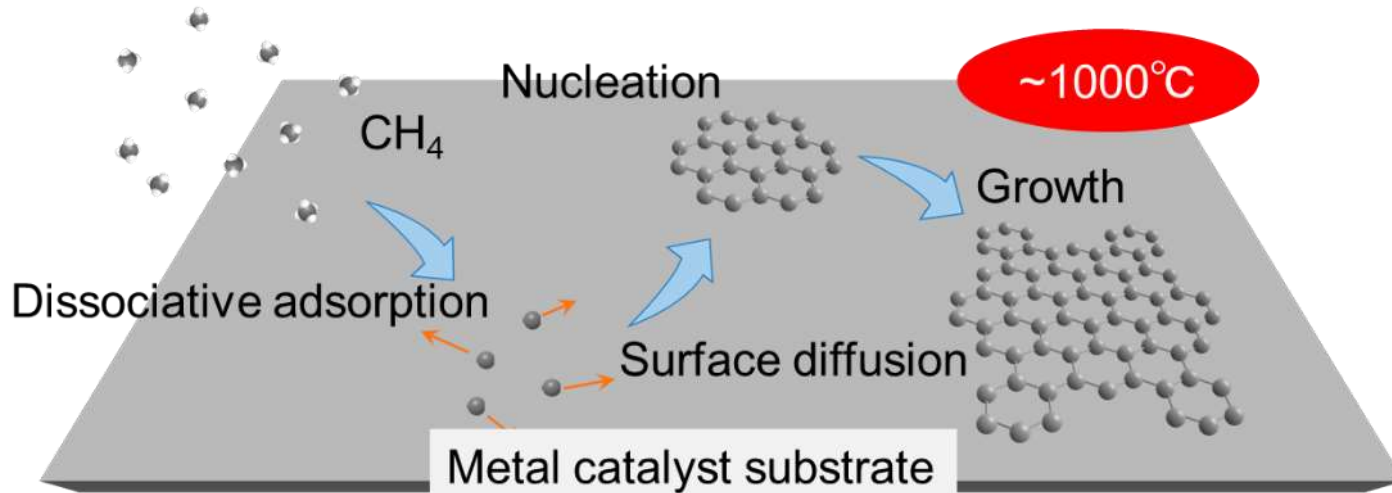
GRAPHENE

2-D honeycomb lattice of carbon atoms



- **Ultrathin ~ 0.3 nm**
- **High Conductivity of Electricity and Heat**
- **High Mechanical Strength**
- **High Optical Transparency (97.7%)**

CVD Growth of Graphene



Graphene prepared by
scotch tape method

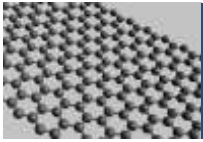


Chemical vapor deposition (CVD)

Large-area and high-quality graphene sheet

Scalable technology

Suitable for industrial applications



CVD Growth on Cu substrate

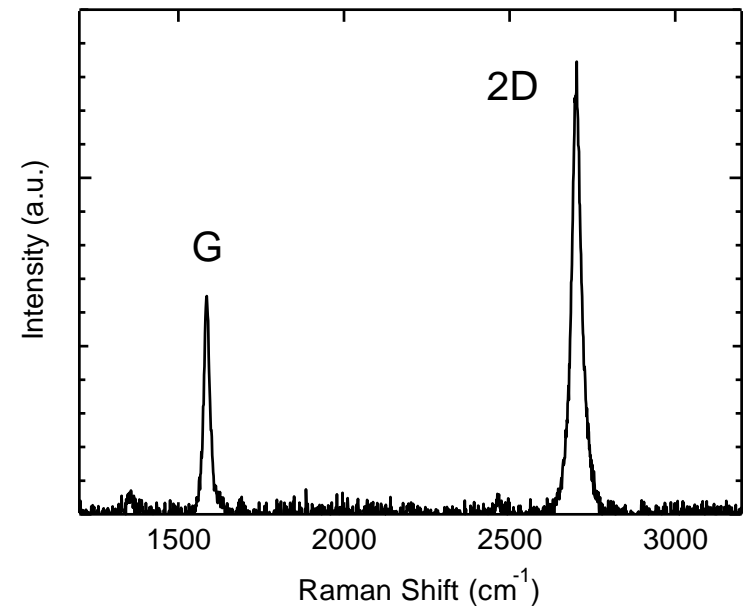
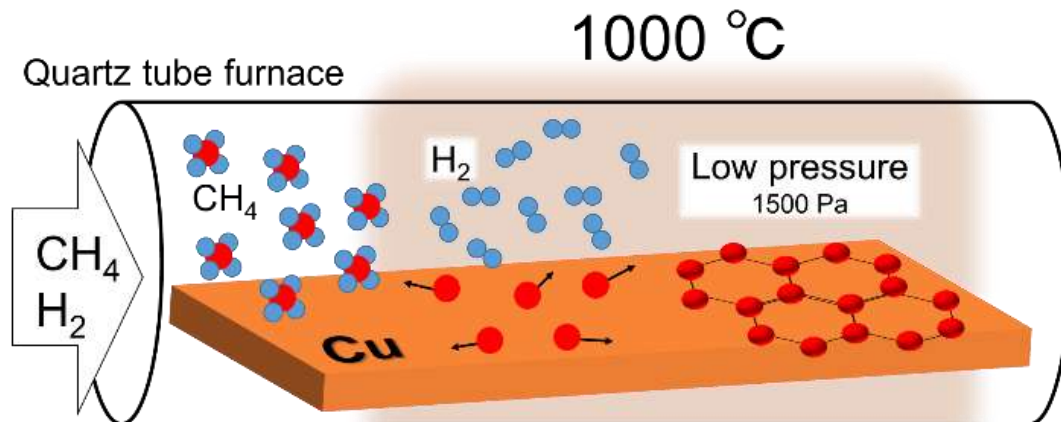
Cu metal catalyst substrate (~ 0.035 mm)

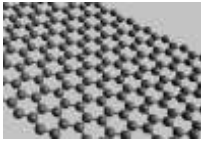
Low cost

Low carbon solubility

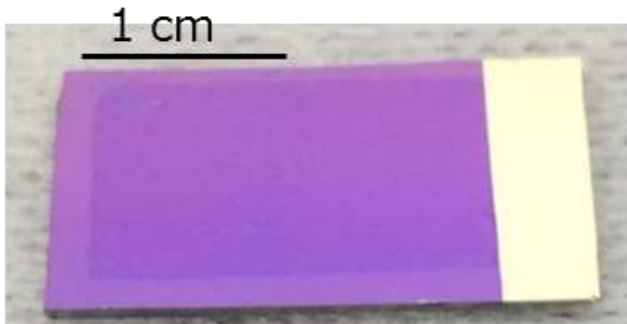
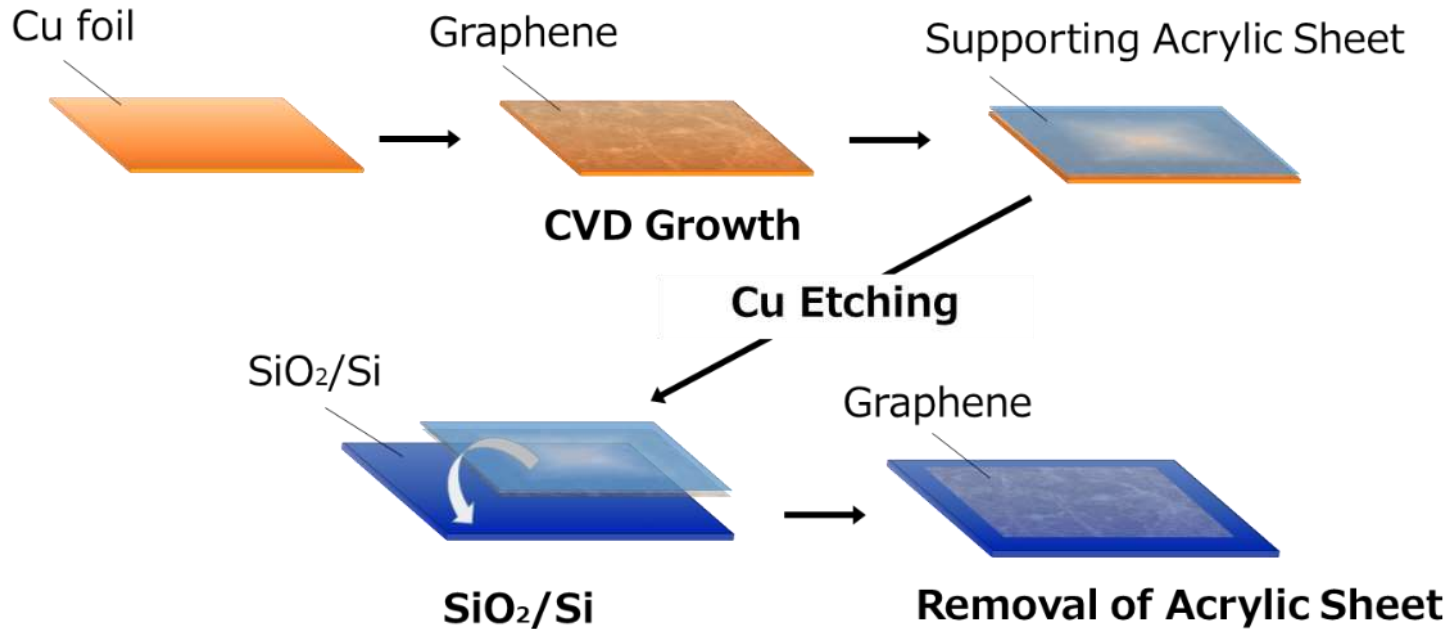
Self-limiting growth of

monolayer graphene





Transfer of Graphene



One-atom-thick graphene transferred onto SiO₂/Si

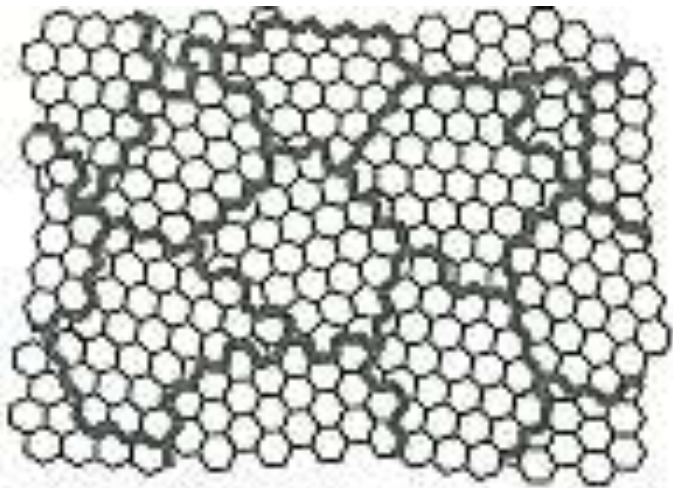
We can transfer graphene onto Various types of substrates.
Quartz glass, PET, PEN, etc.

→ **Device Applications**

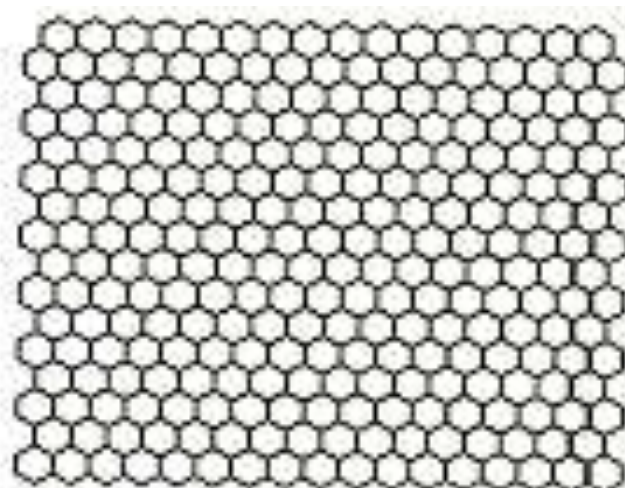


Single Crystalline CVD Graphene

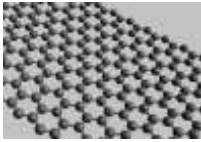
Poly-crystalline graphene
on Poly-crystalline Cu



single crystalline graphene



Single crystalline CVD graphene is favorable to utilize 100% of graphene's properties.

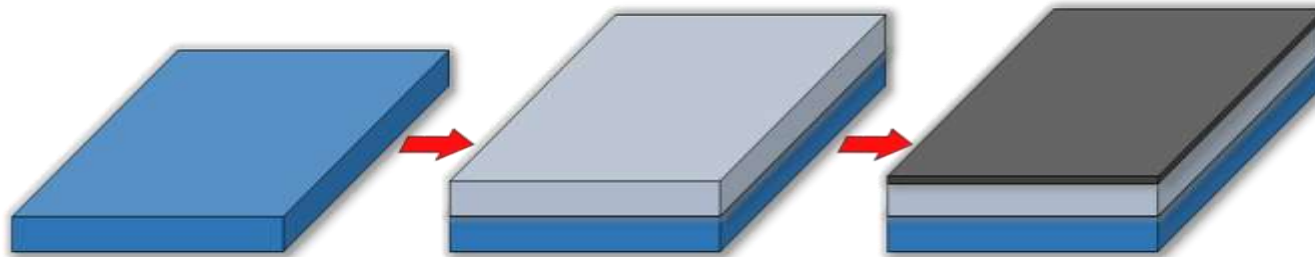


CVD Growth on Iridium

Single crystal sapphire

Epitaxial layer Ir(111)

Single Crystalline Graphene



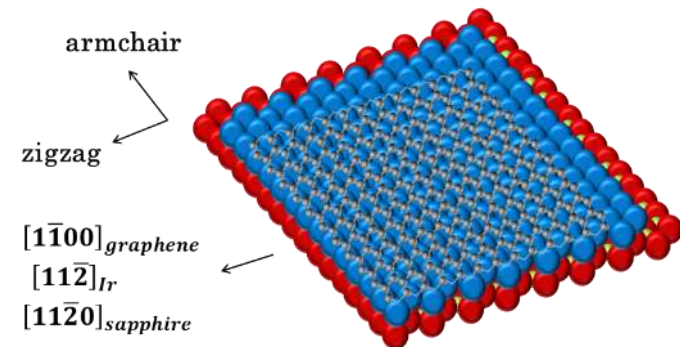
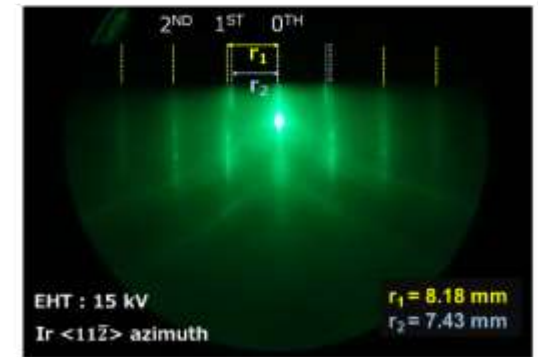
Iridium

- High melting temperature ($\sim 2400^\circ\text{C}$)
- Chemical stability
- Low carbon solubility (0.041 at.%)

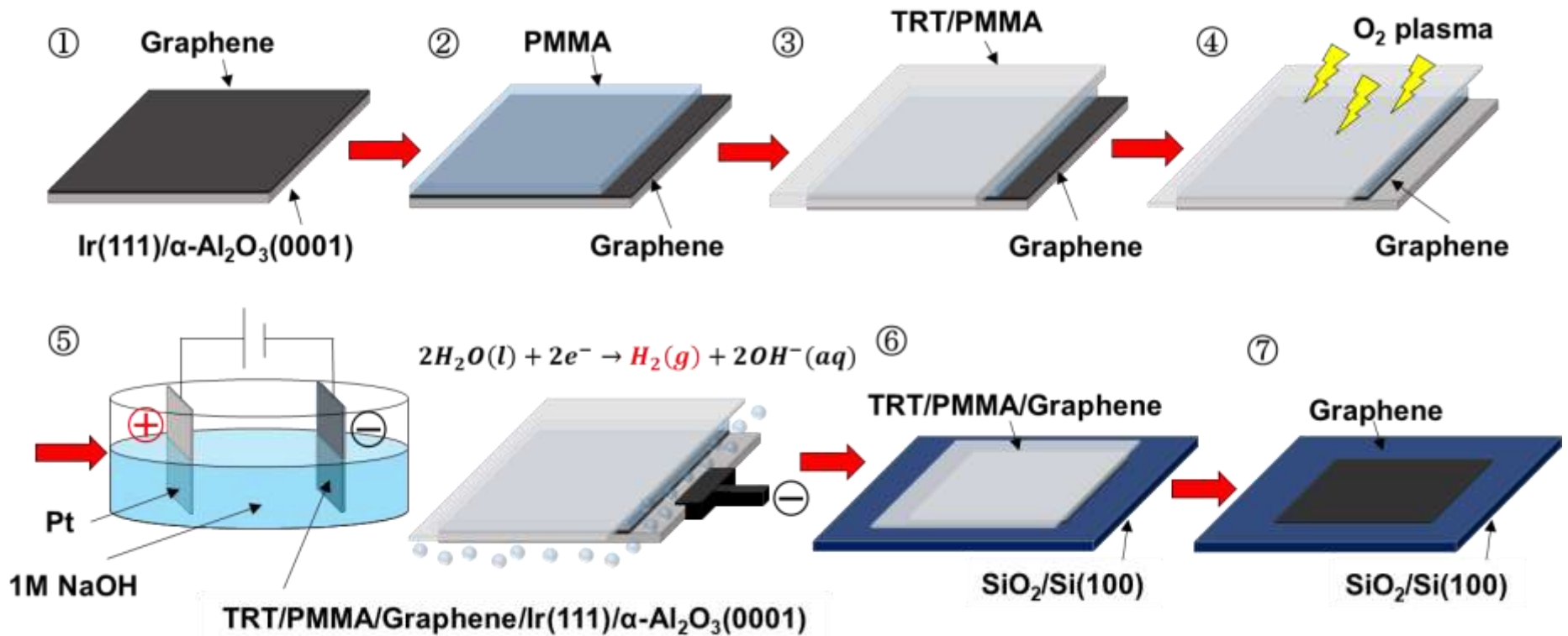
We demonstrated CVD growth of **single-crystalline monolayer graphene on Iridium metal catalyst substrate**

S. Koh, Y. Saito, H. Kodama, and A. Sawabe, *Appl. Phys. Lett.*, **109**, 023105 (2016).

RHEED Pattern for Graphene/Ir(111)

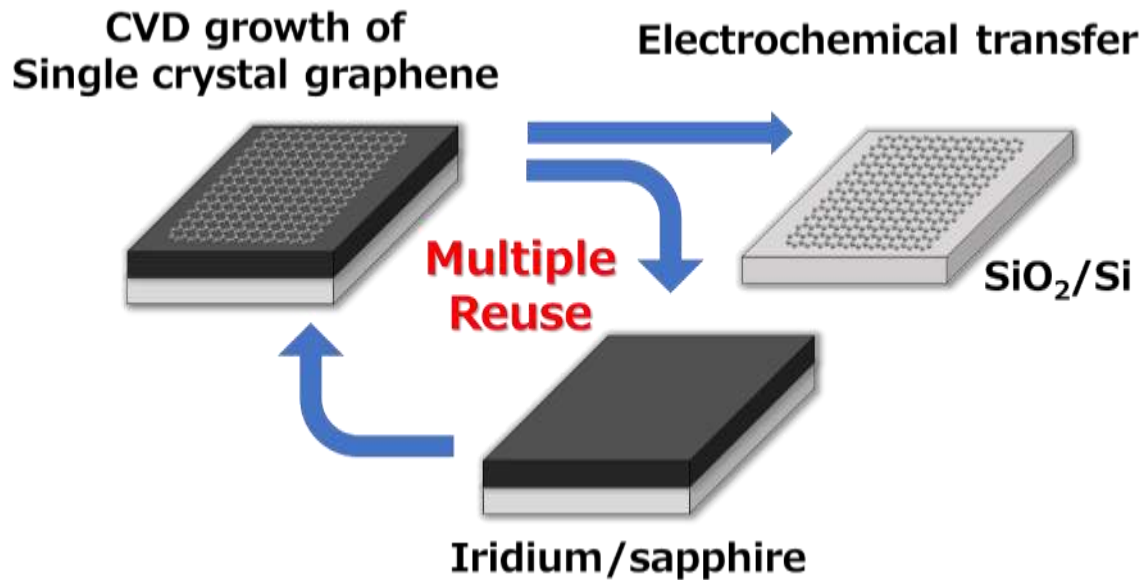


Electrochemical Transfer

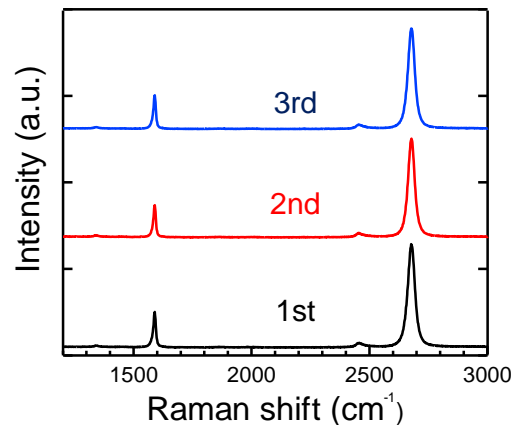
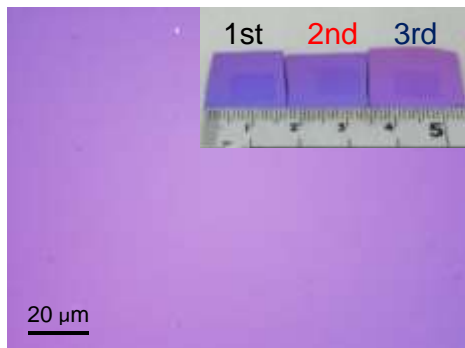


Electrochemical peeling of graphene using H₂ gas bubble generation

Reusability of Ir(111)/Sapphire

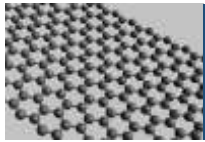


Reusability of Ir/sapphire in multiple cycles of CVD and transfer processes was demonstrated.



S. Koh, Y. Saito, H. Kodama, and A. Sawabe, *Appl. Phys. Lett.*, **109**, 023105 (2016).

A. Sakurai, M. Niki, T. Watanabe, A. Sawabe and S. Koh, To be appeared in *JJAP* (2020)



OUTLINE

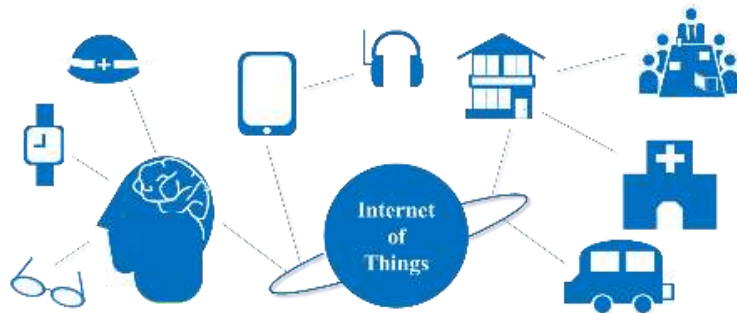
1. CVD growth of graphene on Ir(111)/sapphire
2. Device applications of **CVD graphene grown on Cu**
 - a. **Optically transparent antennas**
 - b. Free chlorine sensors
 - c. Luminescent graphene



Doctor course student
Mr. Shohei Kosuga

Backgrounds: Transparent Antennas

Internet of Things



Various things are connected each other in the network.

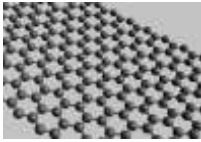
5G system

High frequency ~ 27 GHz

Propagation loss becomes higher.

Propagation distance becomes shorter.

**We need many ANTENNAS!
ANTENNA, ANTENNA, ANTENNA!
Everywhere!**

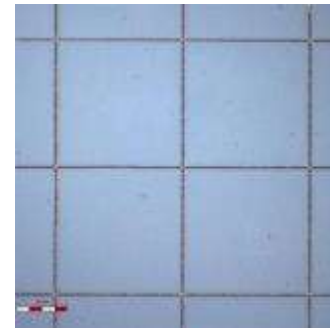


Optically Transparent Antennas

**Optically transparent antennas
maintain transparency of objects
do not change the landscape.**

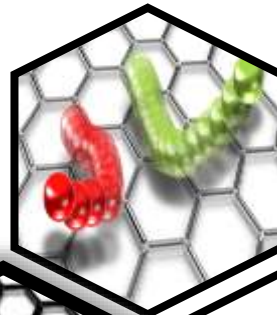


**Metal(Ag) mesh antennas
ITO transparent antennas**



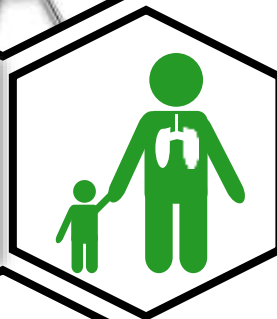
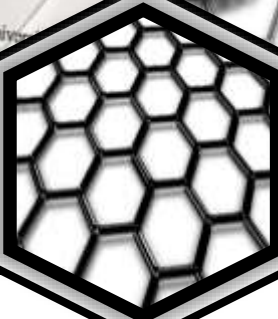
CVD Graphene for Antennas

**High optical
transparency
(~97.7 %)**



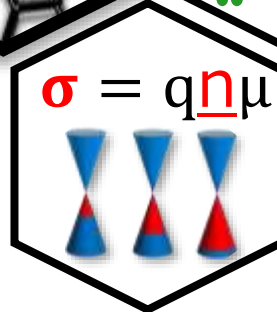
**Excellent
electrical
conductivity**

Flexibility

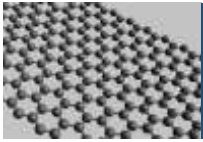


**Metal
free
material**

Ultralight

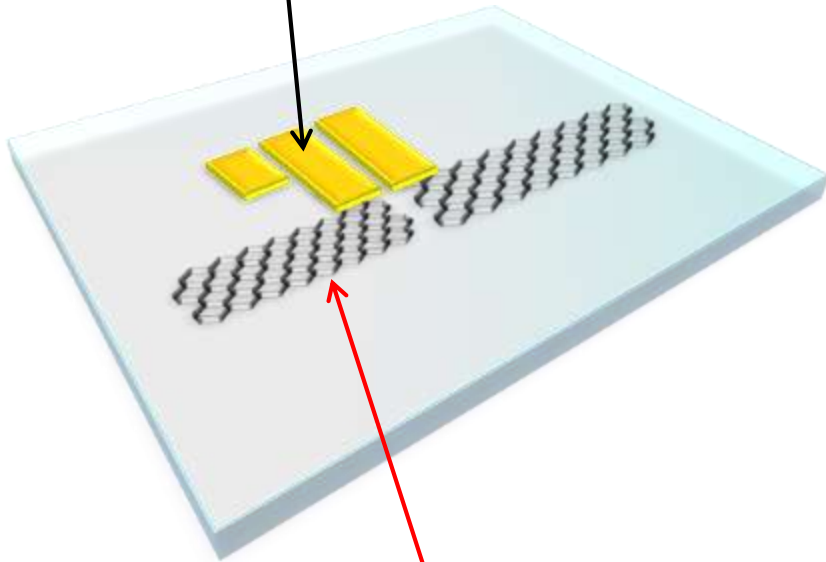


**Tunable
electrical
conductivity**



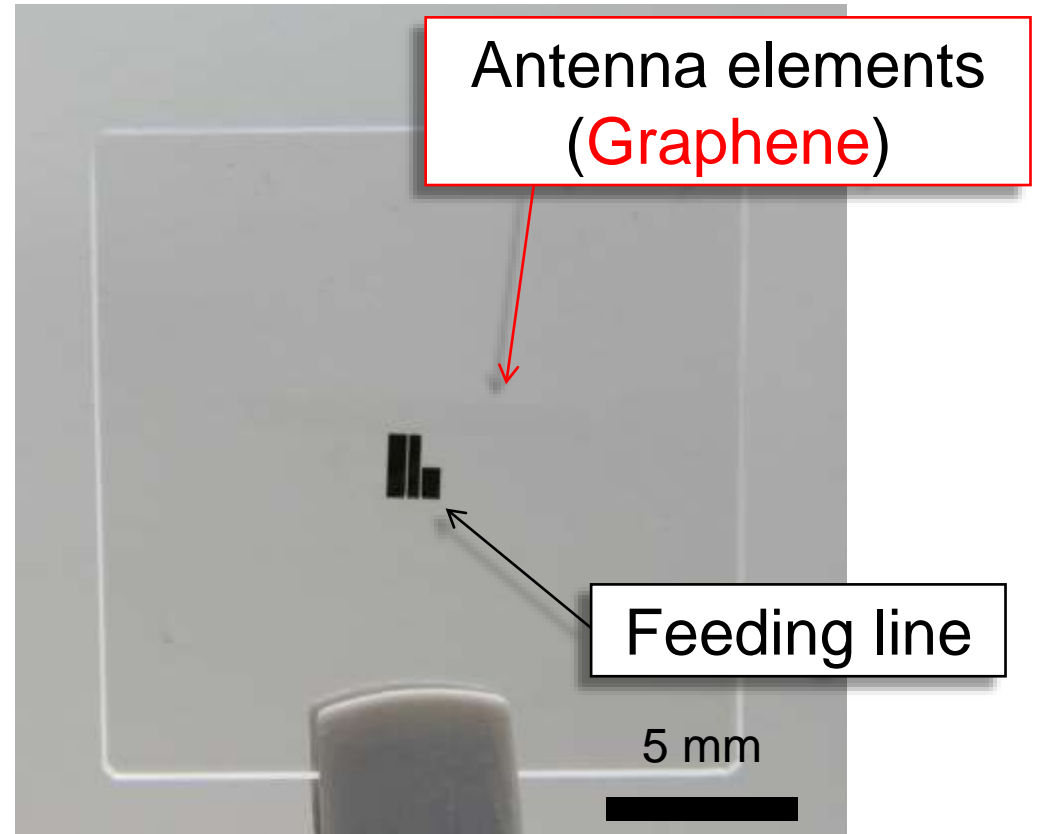
Graphene Transparent Antennas

Au (power feeding)



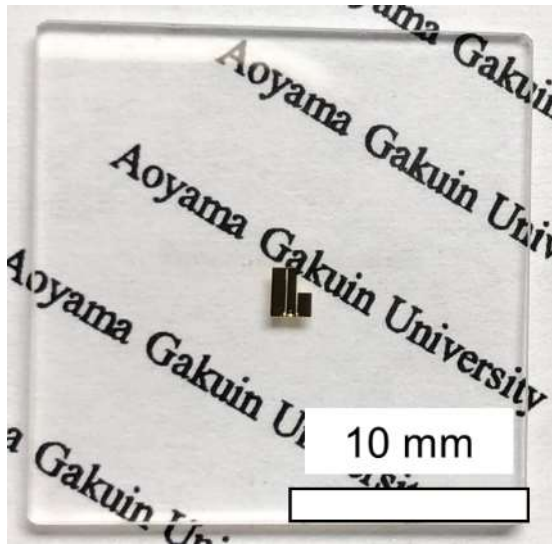
Graphene antenna elements

- Transparency ($\sim 97\%$)
- High conductivity
- Flexibility
- Metal Free



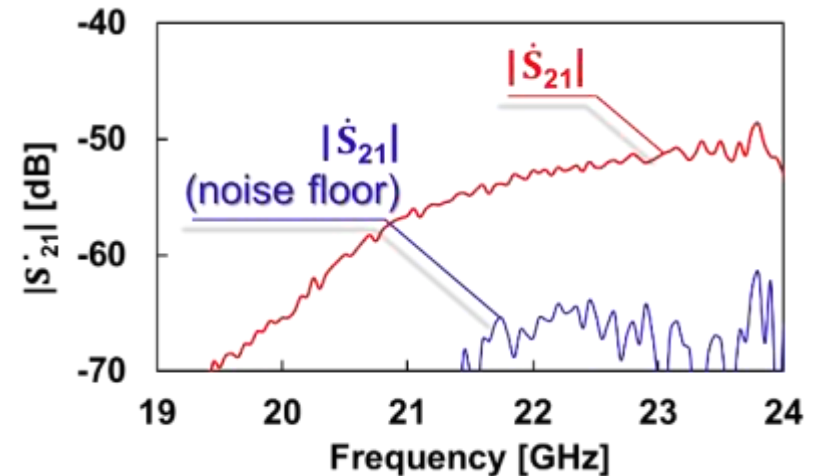
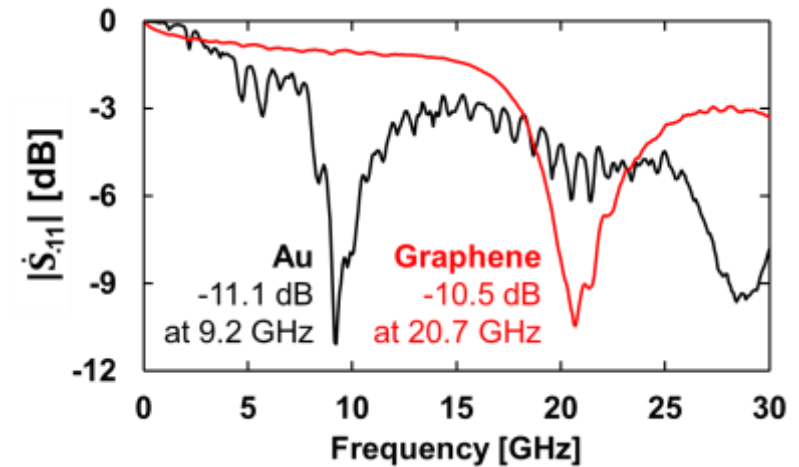
S. Kosuga et al., Appl. Phys. Lett. **110**, 233102 (2017).
S. Kosuga et al., PIERS, Singapore (2017)

Graphene Transparent Antennas



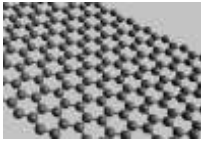
We demonstrated **microwave radiation at ~20 GHz** from monolayer graphene antenna.

Dielectric behavior of monolayer graphene
High Sheet resistance $R_s \sim 700 \Omega/\text{sq}$



S. Kosuga et al., Microwave. Opt. Technol. Lett. **60**, 2992-2998 (2018).

S. Kosuga et al., 30th Asia-Pacific Microwave Conference, Kyoto, Japan, 7 Nov. (2018)



Stacking & Doping

Monolayer



3 layers

Chemical p-doping



Sheet resistivity

700 Ω /sq.



80 Ω /sq.

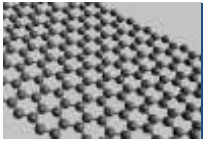
Optical transparency at 550 nm

97.0%



90.6%

**Fabrication of transparent antennas
is now underway.**

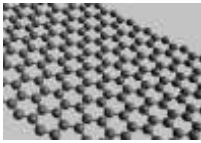


OUTLINE

1. CVD growth of graphene on Ir(111)/sapphire
2. Device applications of CVD graphene grown on Cu
 - a. Optically transparent antennas
 - b. Free chlorine sensors**

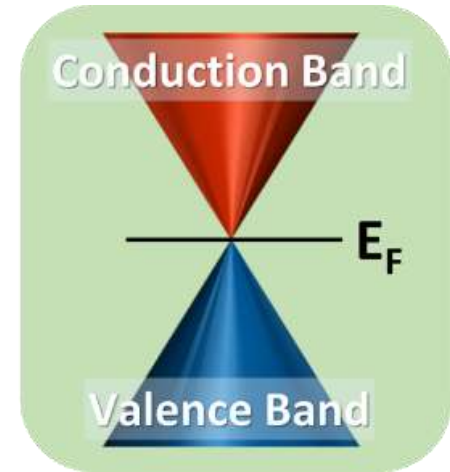
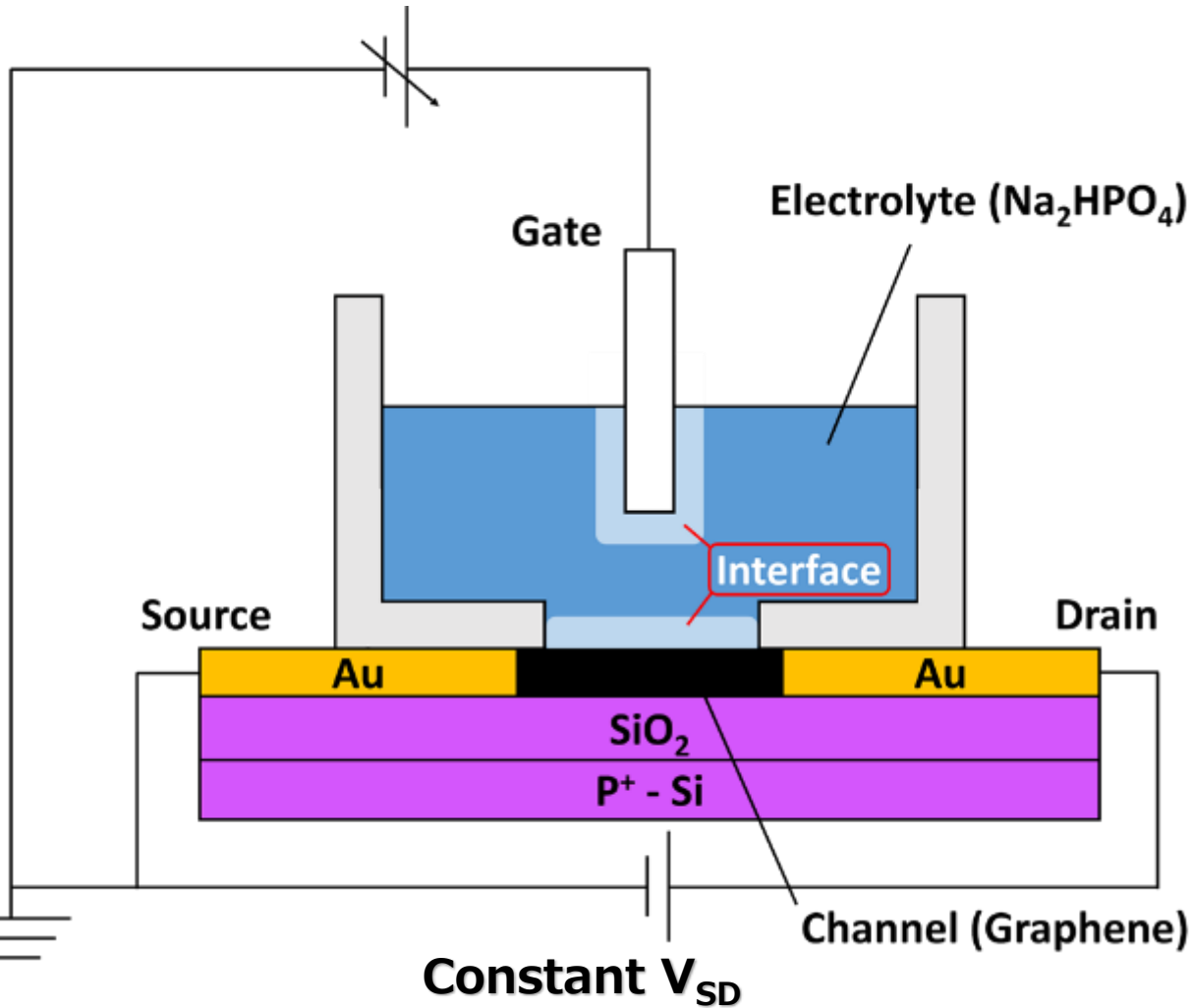


Assistant Professor
Dr. Takeshi Watanabe

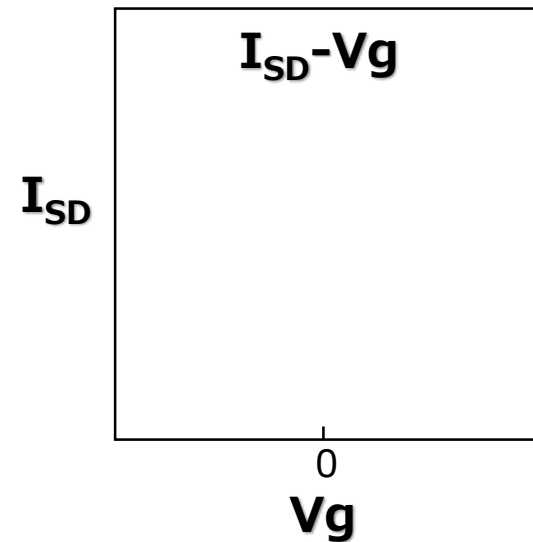


Solution Top-Gated GFET

Conductivity modulation by top-gating

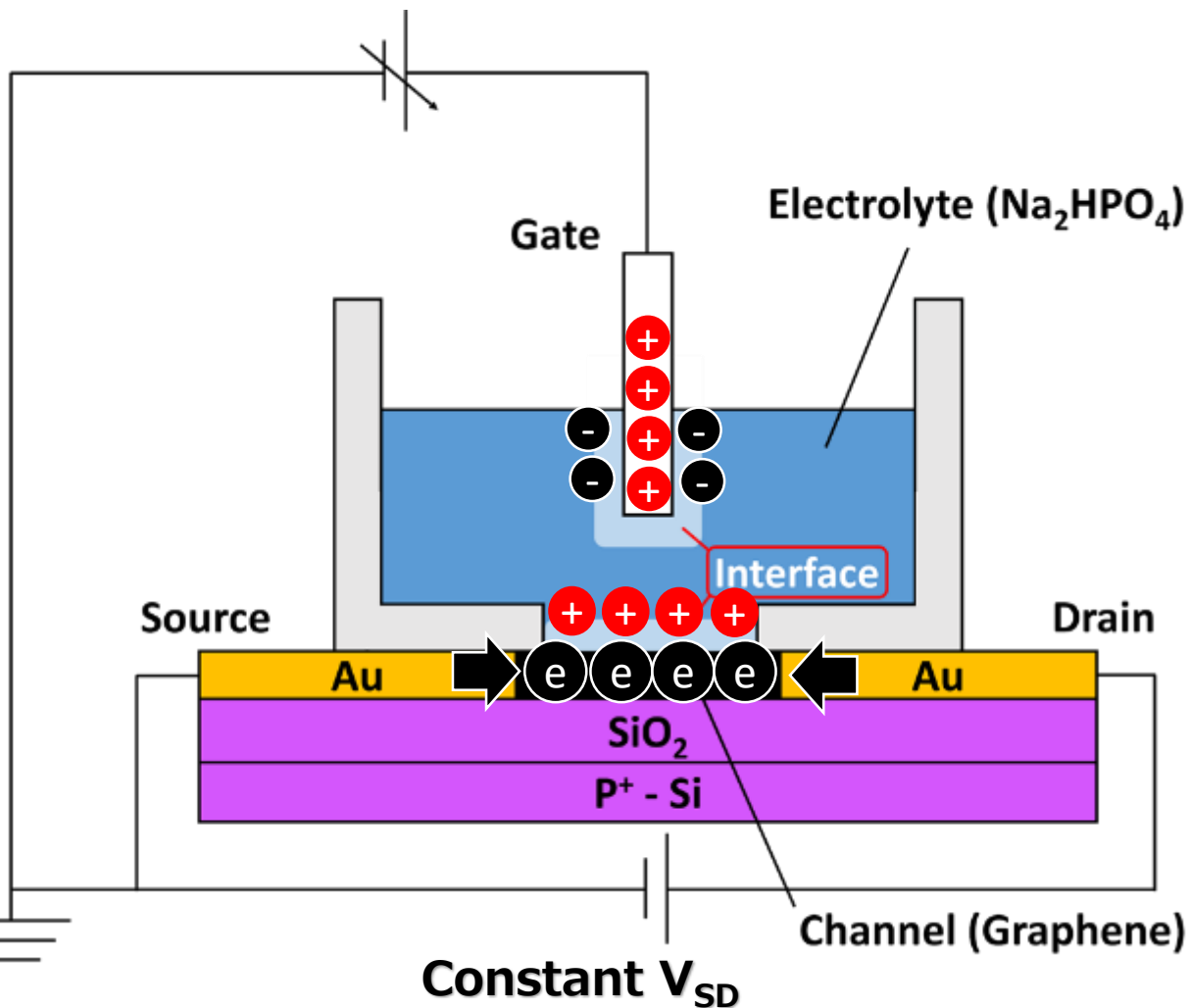


Fermi Level

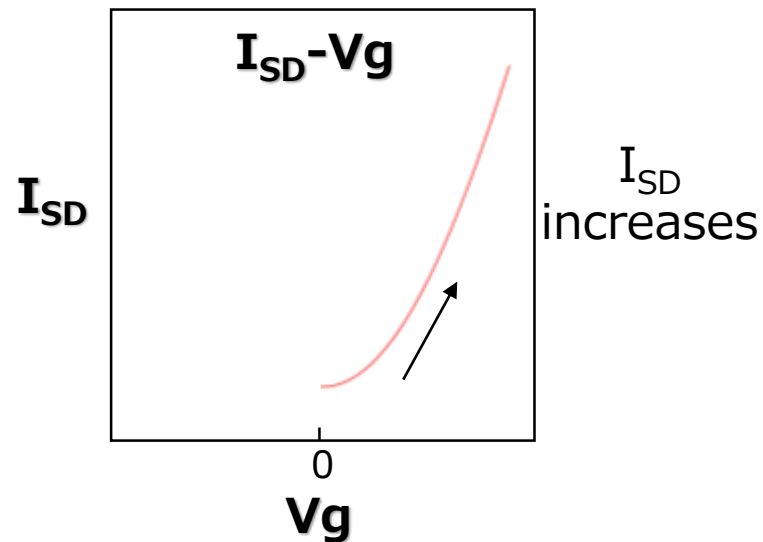
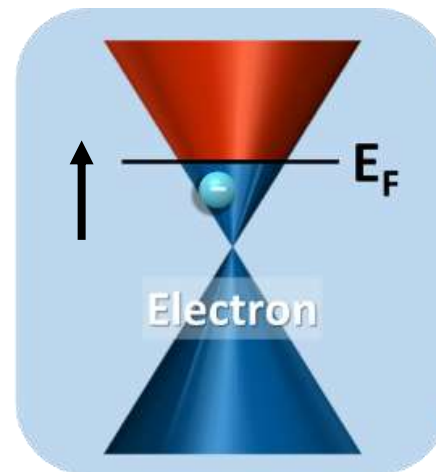


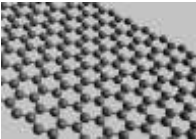
Top-gated GFET in Electrolyte

Positive Gate Voltage V_g



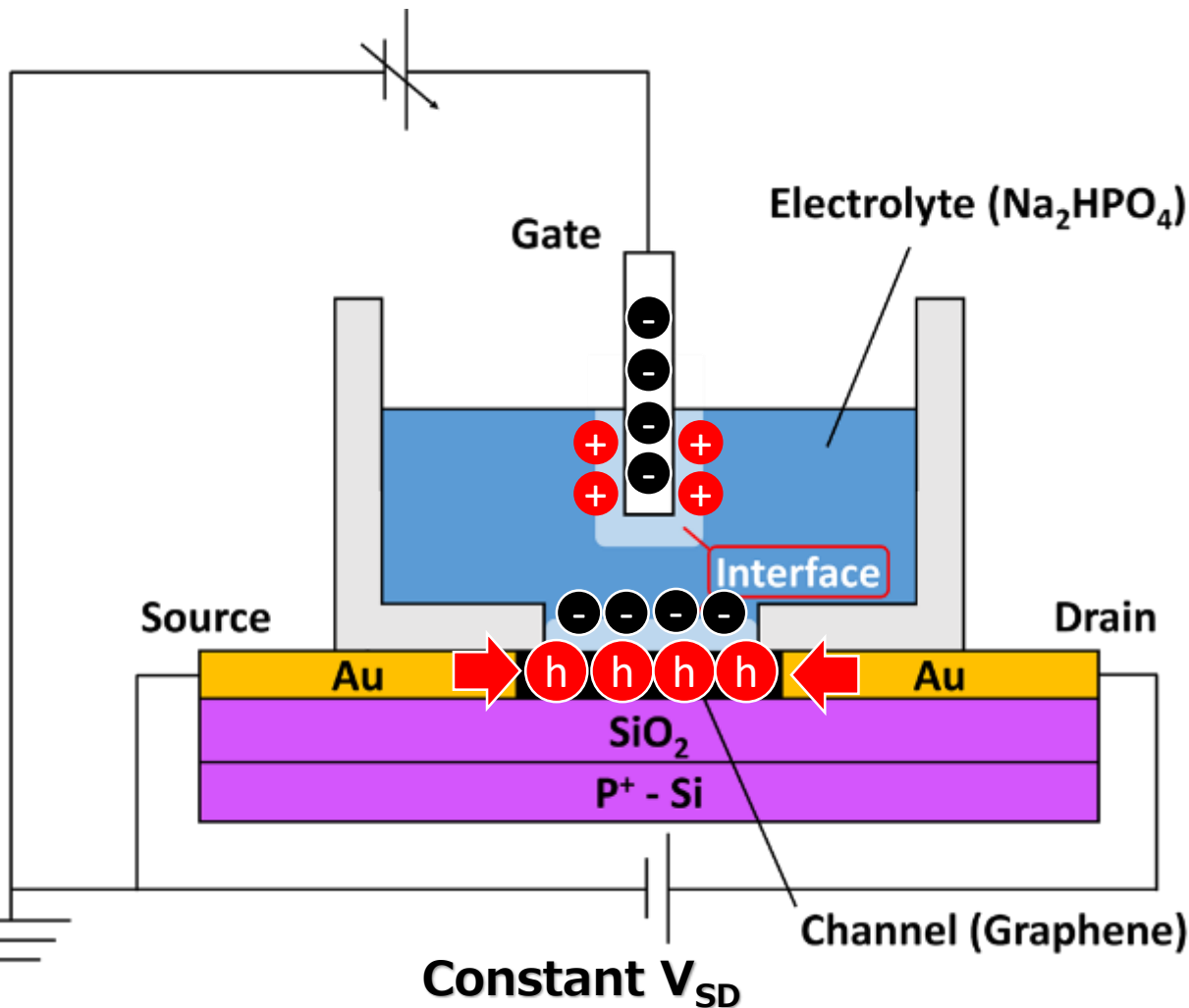
Electron increases (n-type).



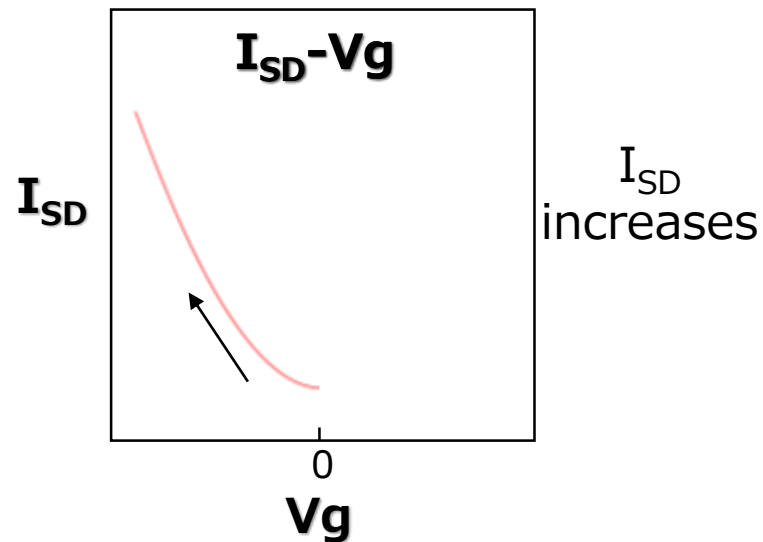
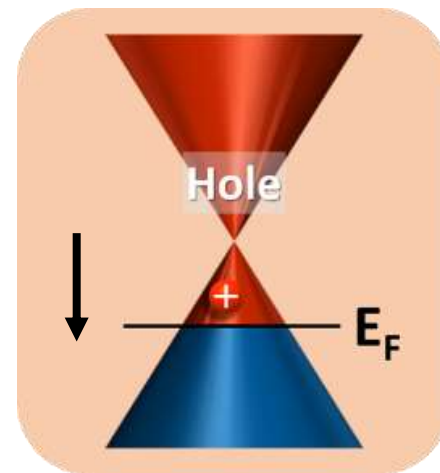


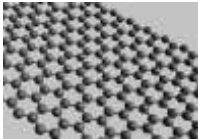
Top-gated GFET in Electrolyte

Negative Gate Voltage V_g

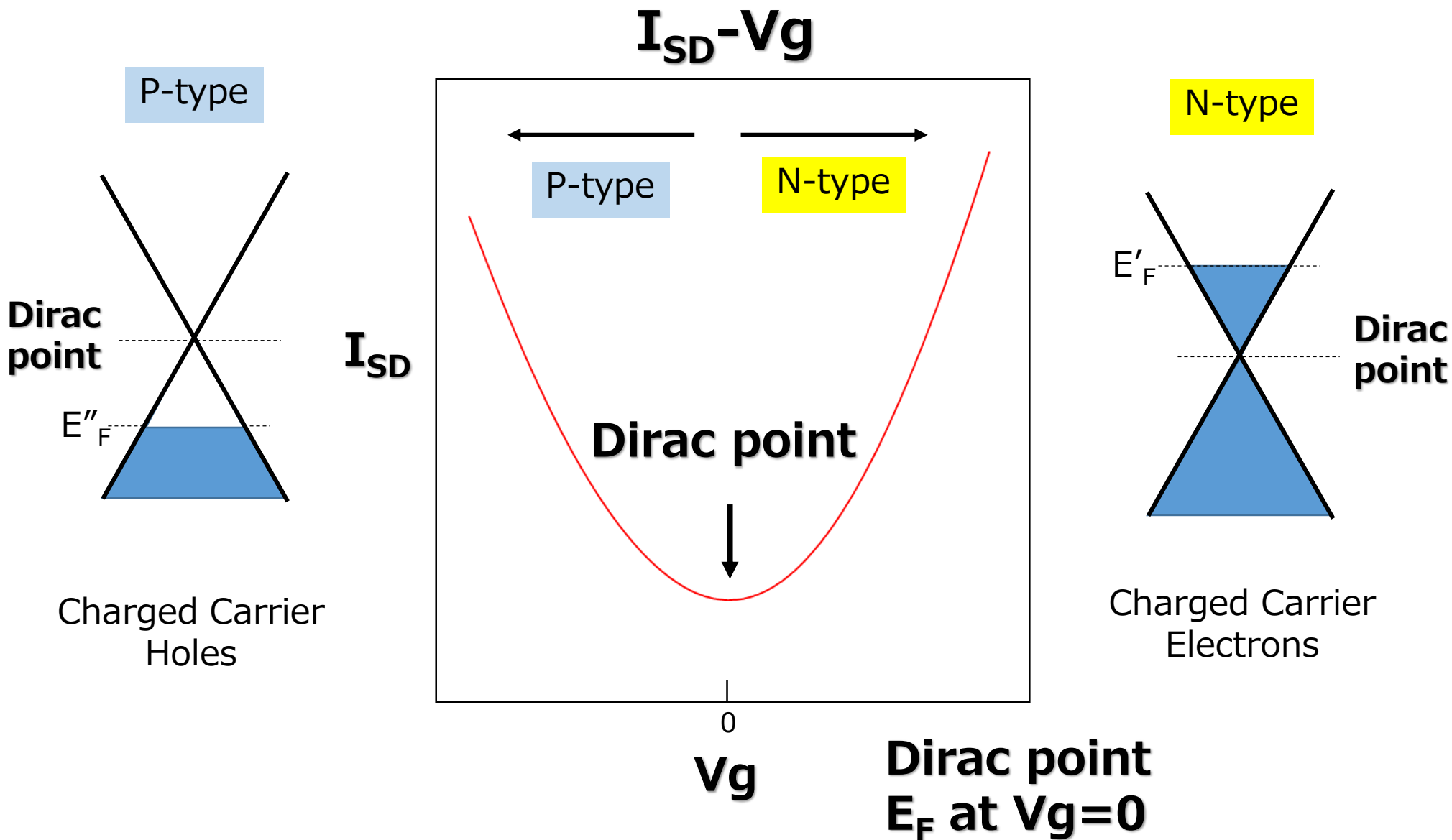


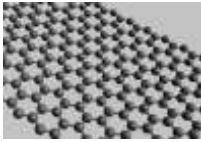
Hole increases (p-type).





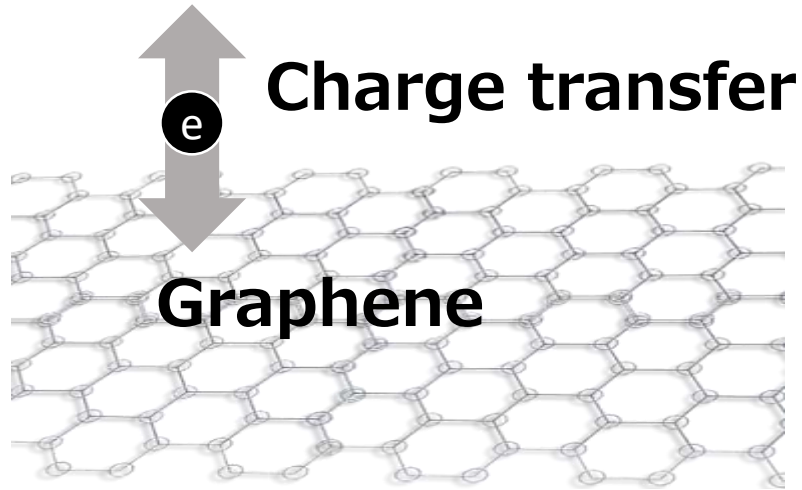
Ambipolar Characteristics of GFET



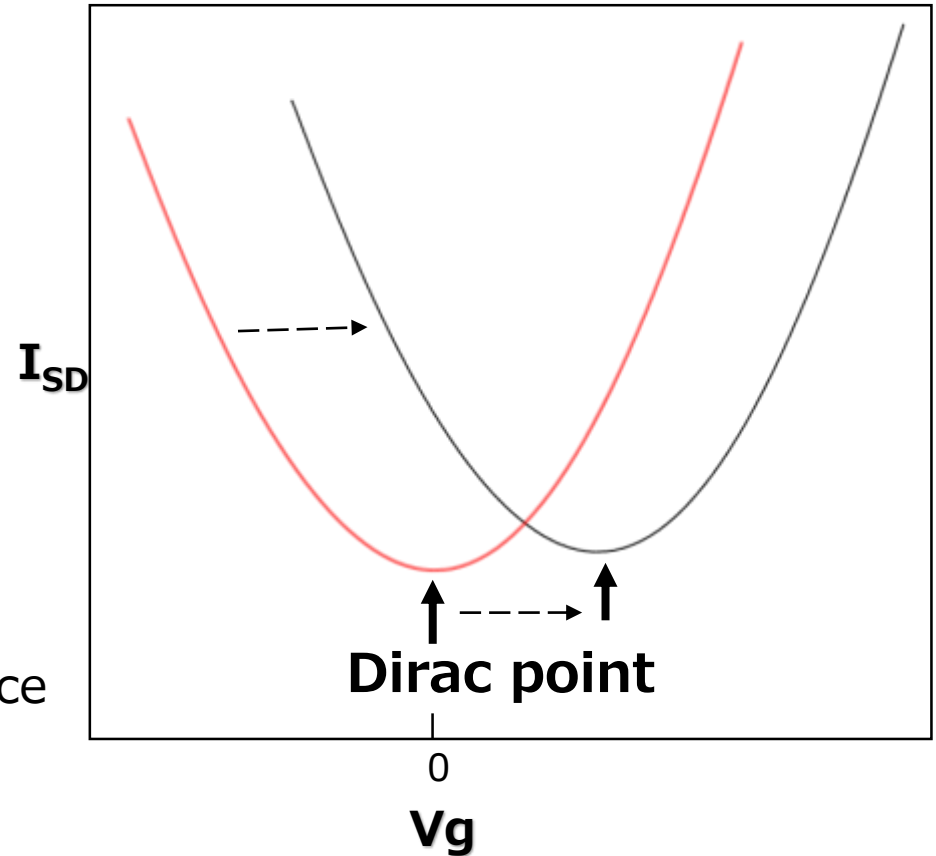
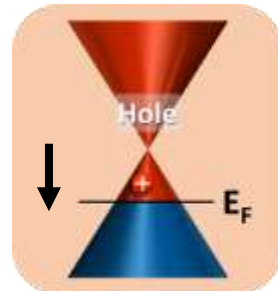
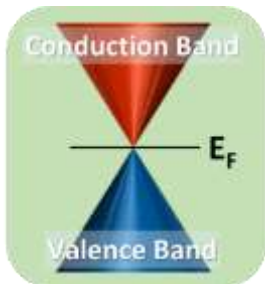


GFET-Based Sensors

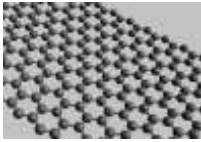
Ions, Molecules etc.



Charge transfer at the graphene surface causes **carrier doping**, resulting in **Shift of Fermi Level**.

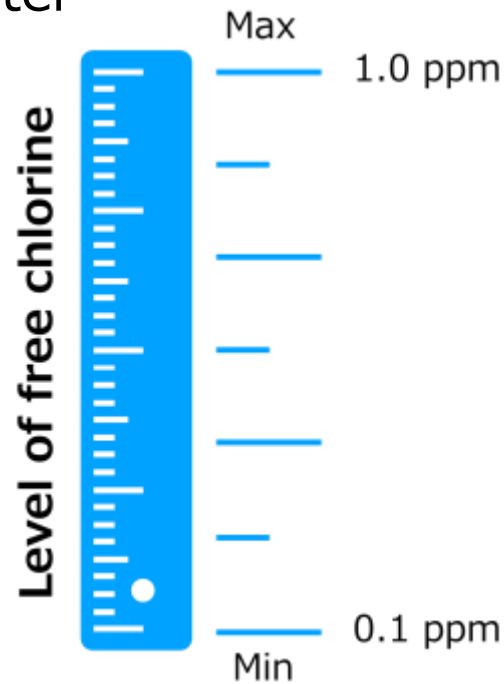


Charge transfer interaction is observed as the shift of $I_{SD}-V_g$ curves.



GFET Free Chlorine Sensors

Chlorine for disinfection of water



Too High

Harmful for human

Too Low

Not effective at killing bacteria and viruses

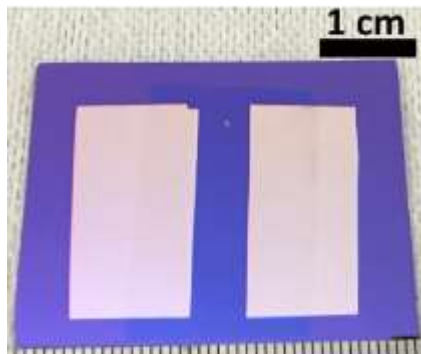
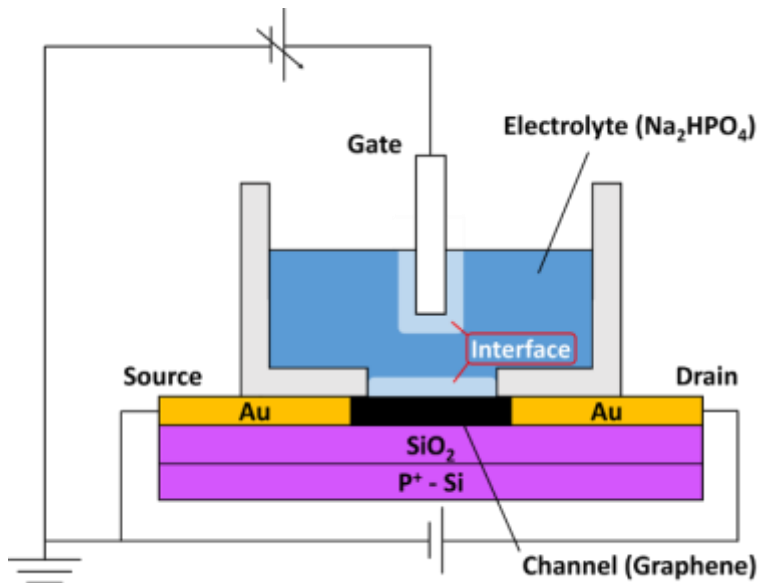
Free chlorine: hypochlorous acid (HOCl) and hypochlorite (OCl⁻) ion

Conventional methods

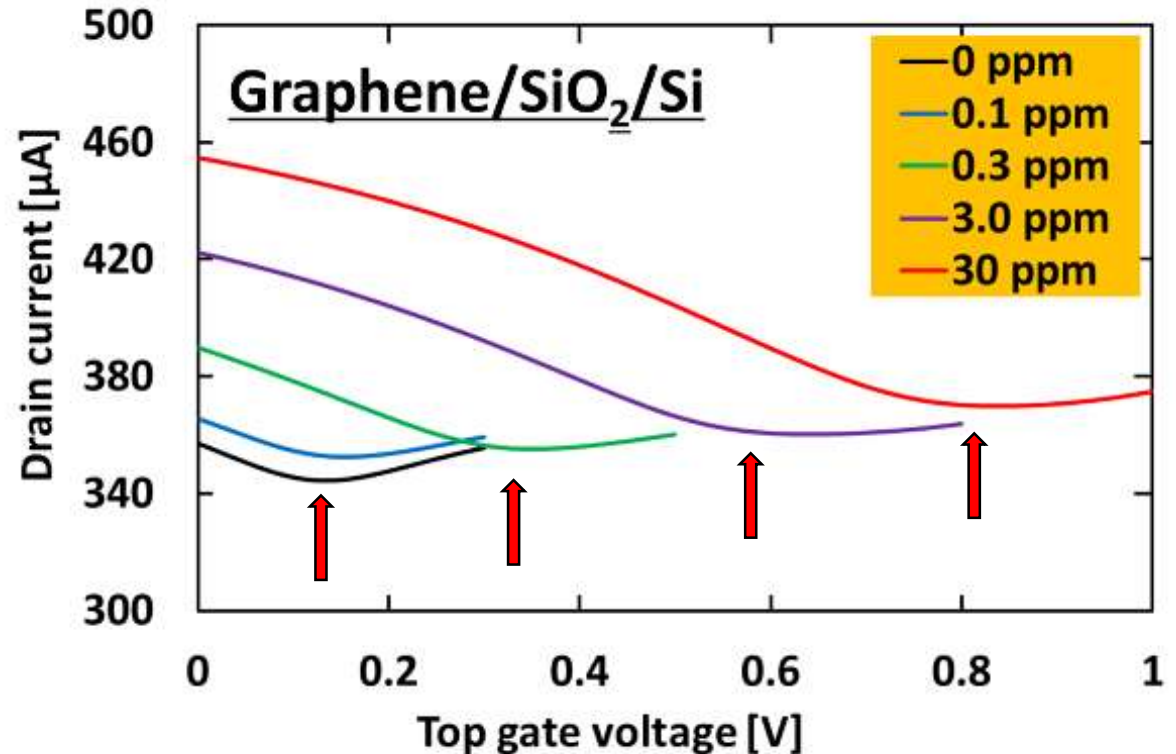
Colorimetric method: Not applicable to continuous measurements

Electrochemical method using Pt: High cost, Oxidation of Pt surface

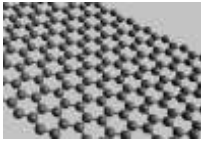
GFET Free Chlorine Sensors



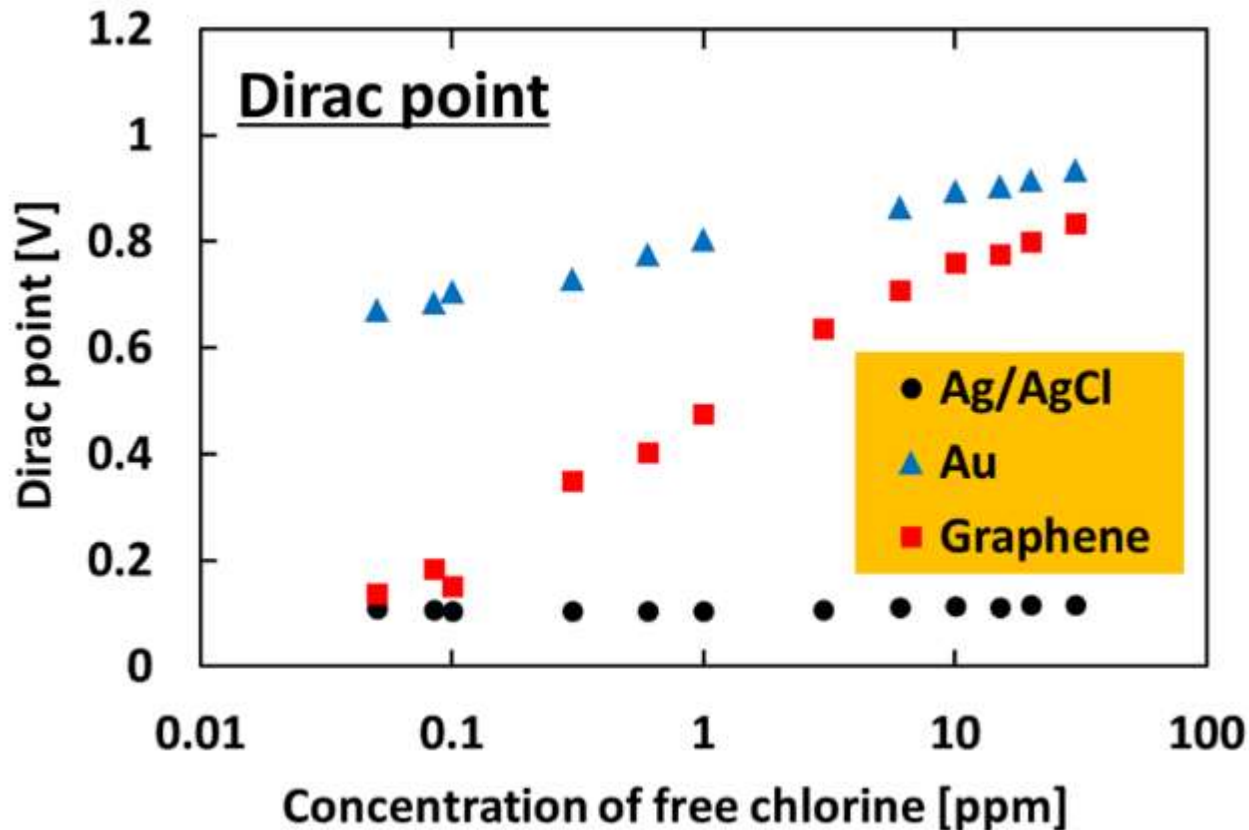
Graphene channel



Dirac point shifts to positive voltage as free chlorine concentration increases.



GFET Free Chlorine Sensors

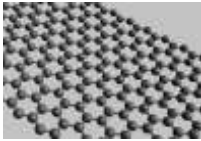


Detection of Free Chlorine Conc. 0.08 ~ 40 ppm

High sensitivity was obtained with

Graphene channel
Graphene top gate

Chlorine sensor.



Summary

1. CVD growth of graphene on Ir(111)/sapphire

CVD growth of single-crystalline monolayer graphene

Reusability of Ir(111) substrates

2. Device applications of CVD-grown graphene

Optically transparent antennas

Free chlorine sensors



Nippon Sheet Glass Foundation for
Materials Science and Engineering

