Exploring spatially and temporally resolved deep-ultraviolet spectroscopy toward understanding and controlling optoelectronic properties of ultrawide bandgap semiconductors

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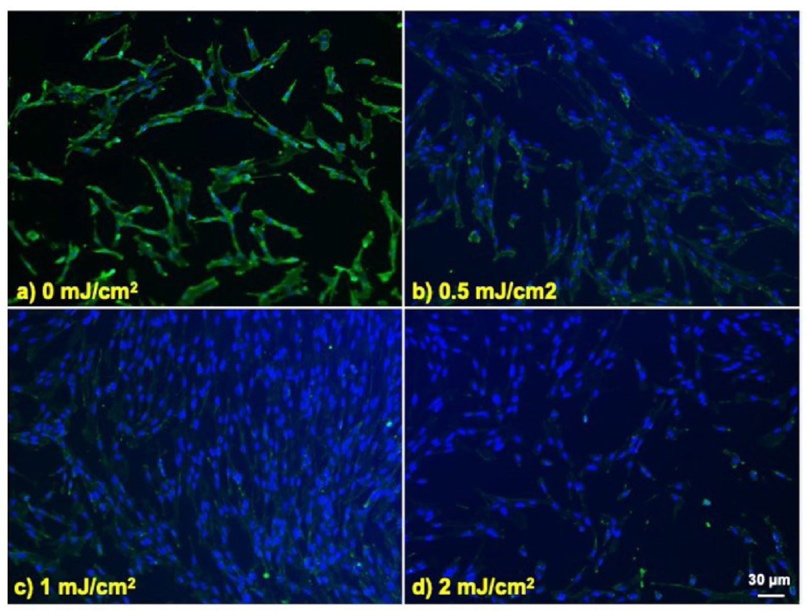
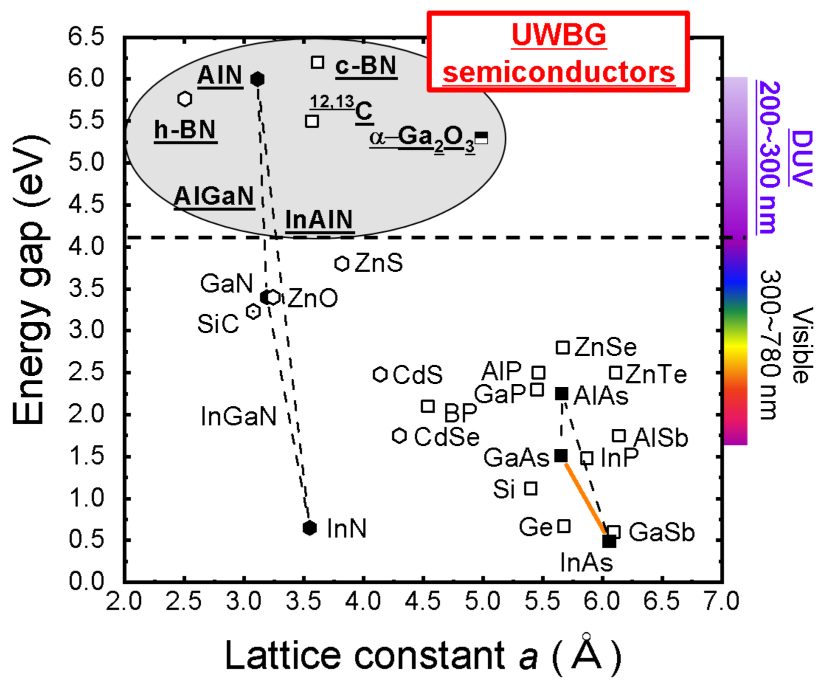
1. Background

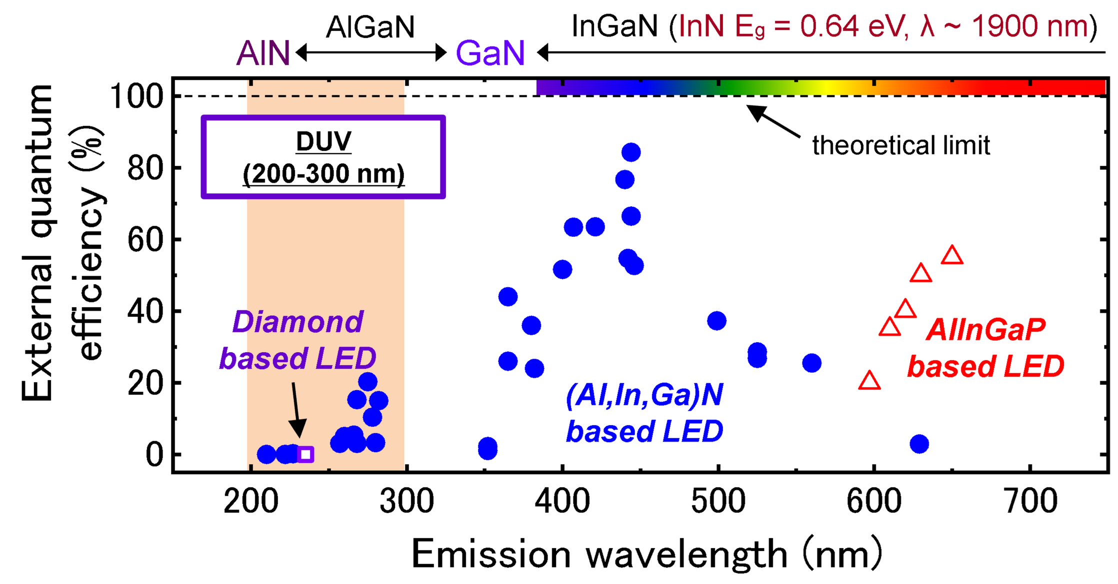
Buonanno *et al.*,
Sci. Rep. **10**, 10285 (2020).



Deep-ultraviolet (DUV) light inactivates influenza and corona viruses.



Ultrawide bandgap (UWBG) semiconductors are the environmentally friendly materials that can generate DUV light.



However, DUV LEDs based on UWBG semiconductors have **low emission efficiency**.

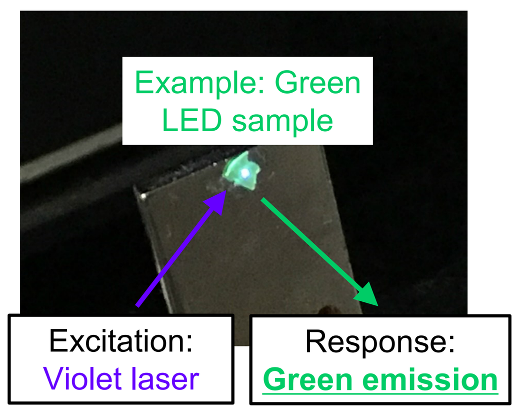
DUV light has attracted much attention for sterilization and water disinfection.

UWBG semiconductors, especially **aluminum gallium nitride (AlGaN)**, are the candidate material of DUV LEDs.

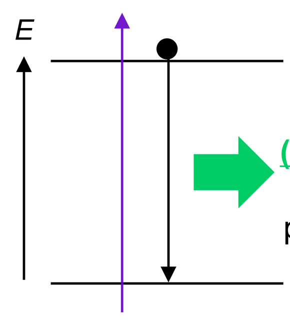
However, the external quantum efficiency (**EQE**) of DUV LEDs is currently very low.

The material physics must be elucidated!

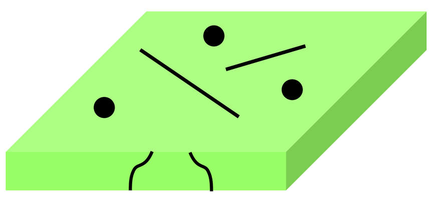
Photoluminescence method



Shorter-wavelength light is externally irradiated



Luminescence (intrinsic response)
→ We discuss the physics of materials!



AlGaN materials have many defects
- point defects (e.g., vacancy)
- line defects (e.g., dislocation)

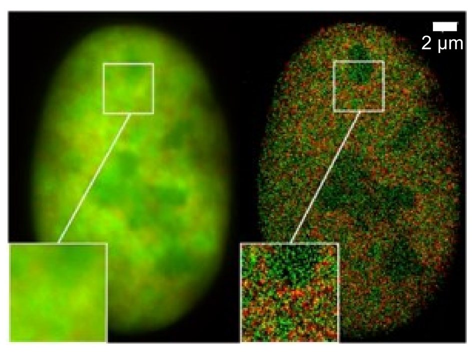
We should elucidate which crystal defects are good/bad defects.

→ **Observe spatially-resolved response with irradiating shorter-wavelength light.**

→ **DUV microphotoluminescence!**

Scanning Near-field optical microscopy (SNOM)

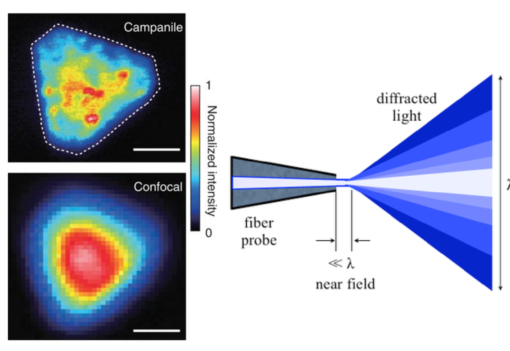
https://commons.wikimedia.org/wiki/File:GFP_Superresolution_Christoph_Cremer.JPG



https://en.wikipedia.org/wiki/Near-field_scanning_optical_microscope

Diffraction limit of light
Conventional (**far-field**) optical microscope cannot visualize sub-wavelength structures with lateral resolution greater than 200 nm.
But, spectroscopic information is invaluable for material characterization.

Scanning probe microscope and electron microscope can visualize nanometer structures.
But, rather difficult to obtain spectroscopic information.



SNOM
Detect **near-field** information by scanning a SMOM probe (e.g., optical fiber probe).

Combination of optical microscope and scanning probe microscope
⇒**Spectroscopic information with high spatial resolution**

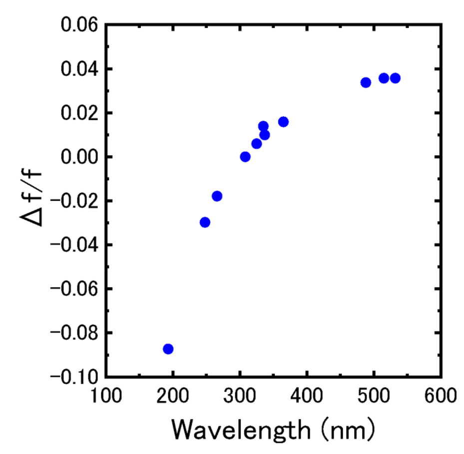
History of DUV-SNOM

References	Excitation source	Mode	Resolution	Experiment, samples
Sands *et al.*, J. Raman. Spectrosc. **33**, 730 (2002).	Ar+ SHG (244 nm)	I mode	200 nm	Raman signals, diamond
Aoki *et al.*, APL **84**, 356 (2004).	YAG 4HG (266 nm)	I mode	50 nm	PL signals, organic materials
Pinos *et al.*, APL **95**, 181914 (2009).	EL (285 nm)	C mode	150 nm	EL signals, AlGaN LED
Pinos *et al.*, JAP **109**, 113516 (2011).	Ti:S THG (258 nm)	I-C mode	100 nm	PL signals, $Al_{0.3-0.5}GaN$ layers
Marcinkevicius et al., APL **105**, 241108 (2014).	Ti:S THG (**227 nm**)	I-C mode	100 nm	PL signals, $Al_{0.6-0.7}GaN$ layers

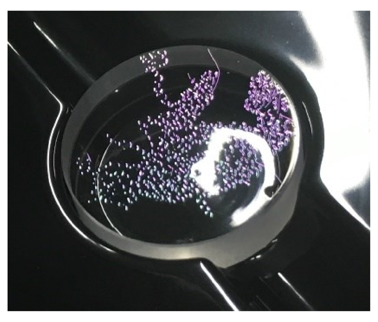
Previous DUV-SNOM cannot measure DUV LEDs emitting below 240 nm※.
※which safely inactivate influenza and corona viruses.

The purpose of the research 1
1. **Development of DUV-SNOM operating at the shortest wavelength**
2. **Visualizing the luminescence of DUV LEDs emitting below 240 nm**

Problems of DUV microspectroscopy

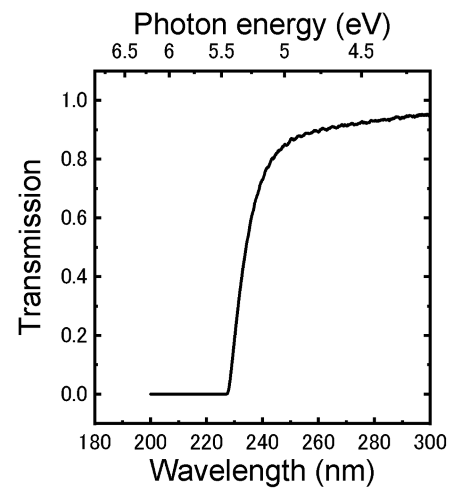


DUV refractive lenses have **huge chromatic aberration**



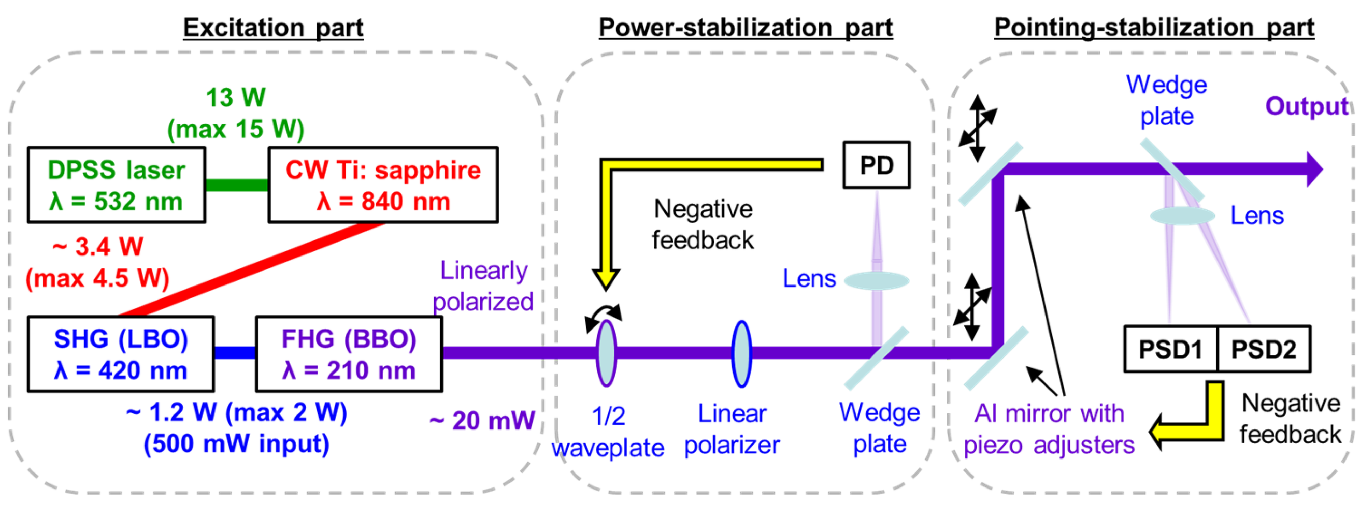
Optics are easily damaged by DUV irradiation
(**power and pointing instability**)

Limited choice of excitation sources

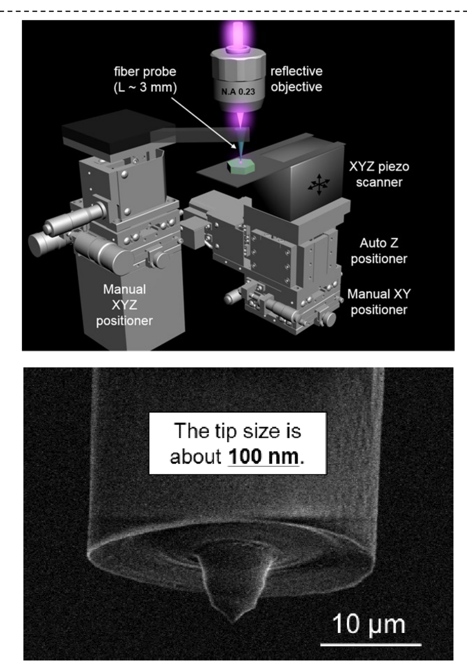


Optical-filter technology is immature
(**no commercial edge filters**)

Our solutions

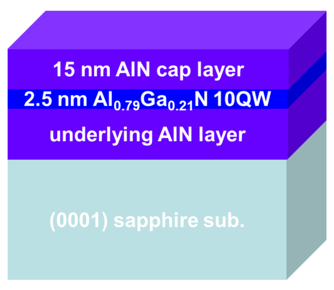


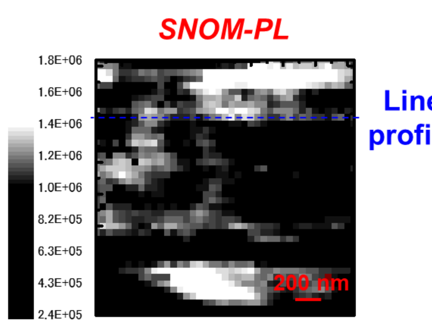
Fourth harmonics of **CW** Ti: sapphire laser
Development of **power- and pointing-stabilizing system**

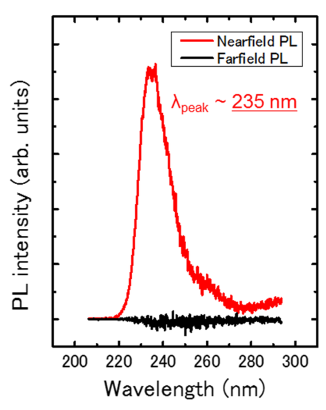


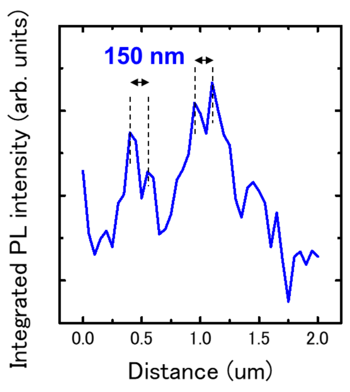
Reflective objective system
Low-loss (~3 mm long) SNOM probe

Result 1









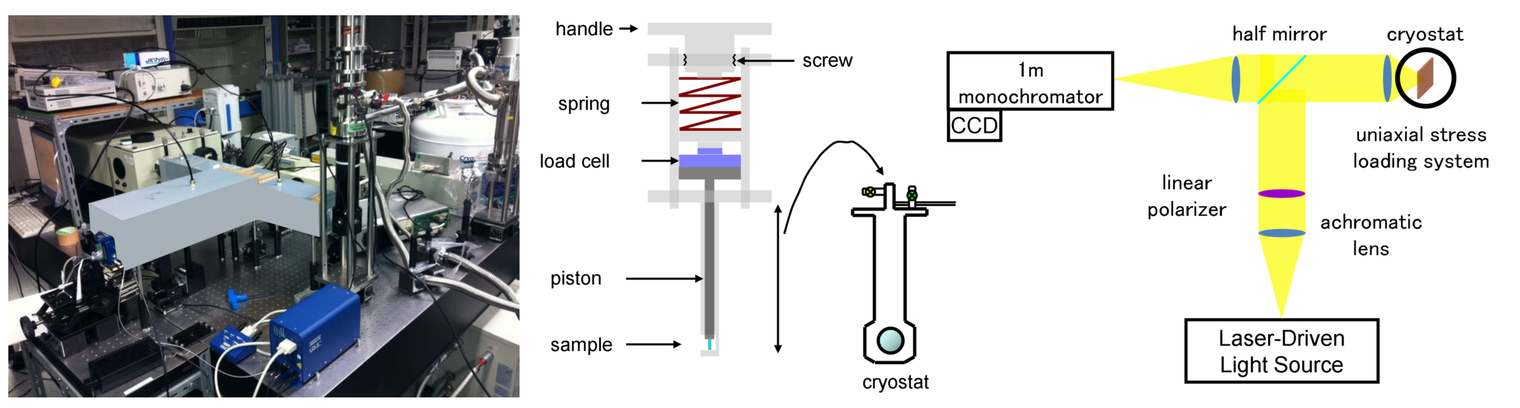
With **210 nm** excitation wavelength, **below 240 nm emissions** from AlGaN QWs can be measured.

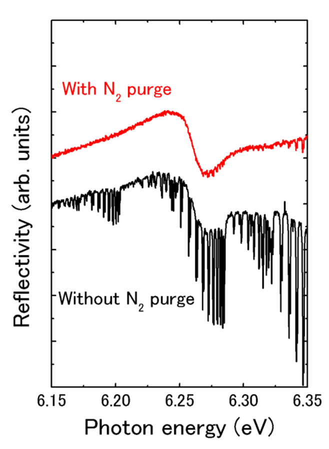
The lateral resolution is greater than **150 nm.**
(beyond the diffraction limit of light)

Screw dislocations act as **radiative** recombination centers.

Ishii *et al.*, APL Photon. **4**, 070801 (2019).
"**Featured**" and "**Scilight**" article

Research 2:
DUV reflectance spectroscopy under uniaxial stress



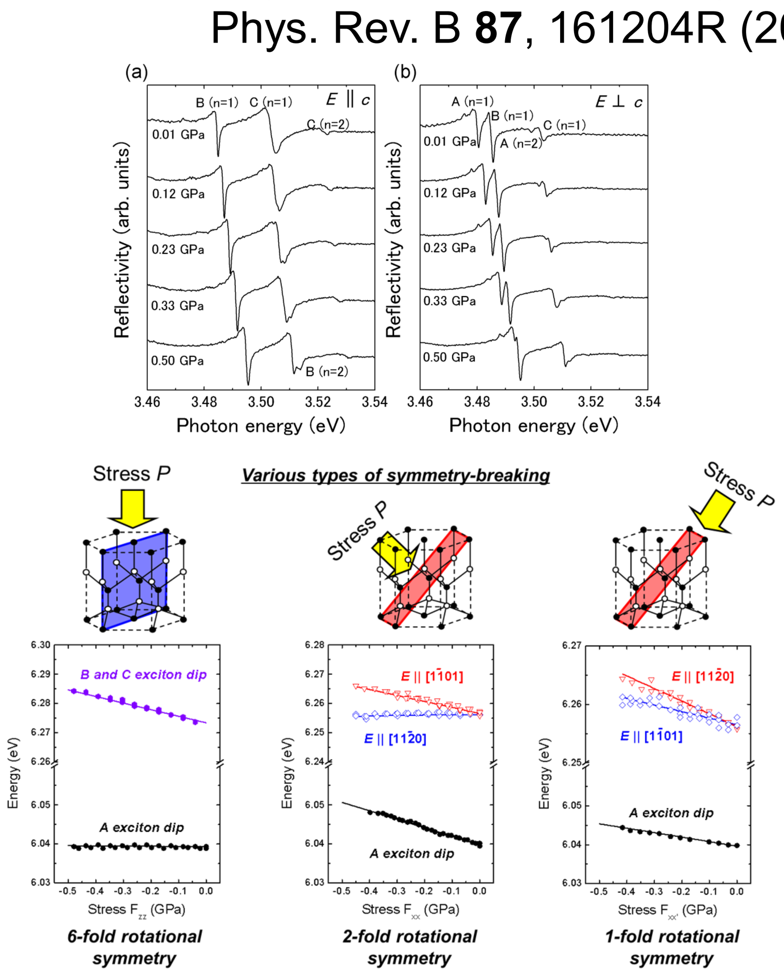


The purpose of the research 2
Reveal the exciton fine structure of aluminum nitride (AlN) and determine the deformation potentials.

We constructed a DUV reflectance spectroscopy system that can apply uniaxial stress at cryogenic T.
Optical path is fully purged by nitrogen to reduce oxygen light-absorption.

Result 2

Ishii *et al.*, Phys. Rev. B **81**, 155202 (2010) for GaN,
Phys. Rev. B **87**, 235201 (2013) for AlN.
Phys. Rev. B **87**, 161204R (2013) for AlN.



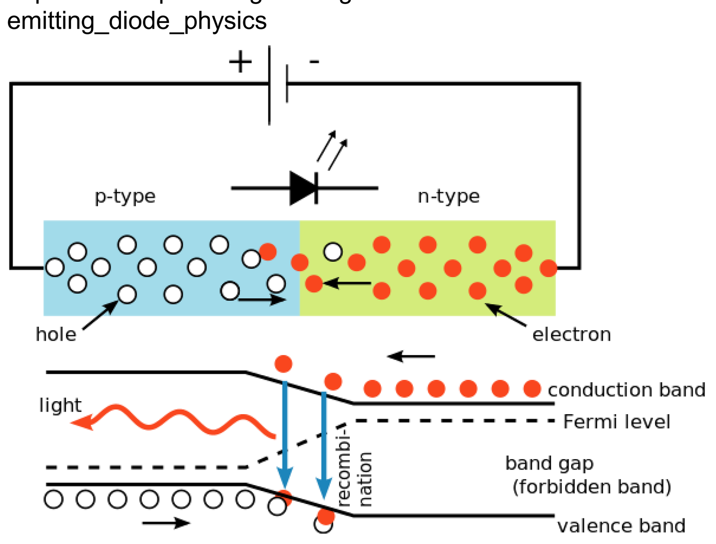
Applying uniaxial stress significantly changes the optical response of GaN and AlN.

We determined all the deformation potentials of AlN and proposed the exciton fine structure.

These knowledge enables to **predict** the device characteristics of DUV light-emitting devices.

Research 3:
Toward p-type electric conductivity control of AlN

https://en.wikipedia.org/wiki/Light-emitting_diode_physics

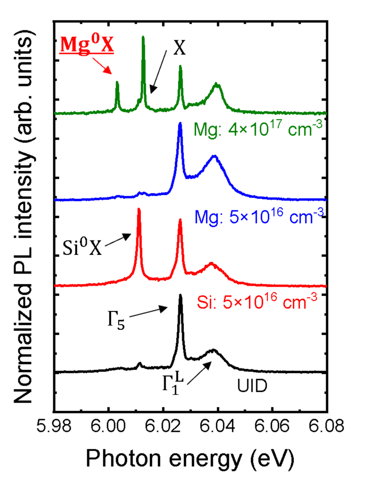


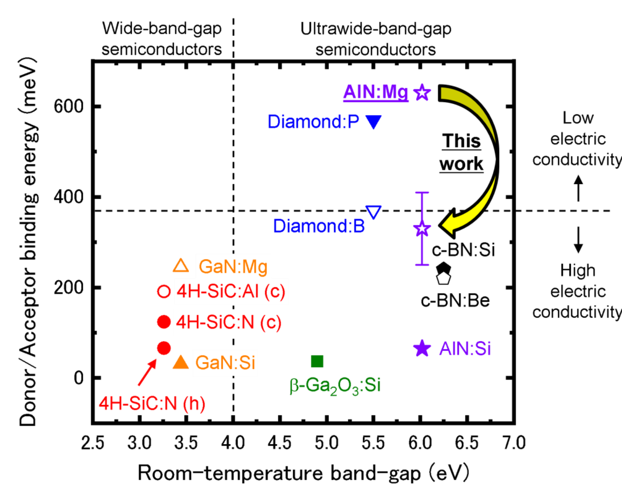
Transparent p- and n-type layers are required to realize efficient LEDs.

However, p-type electric conductivity control of AlN by Mg-doping was believed to be **unfeasible**.

Result 3

Ishii *et al.*, Phys. Rev. B **108**, 035205 (2023).





Our high-resolution DUV photoluminescence spectroscopy deduced an **unexpectedly small** Mg acceptor binding energy of AlN.

We gave a novel promising method for realizing efficient DUV LEDs!