Development of high radiation tolerance detector with CIGS

K. Itabashi[A], M. Togawa[A], J. Nishinaga[B], T. Isobe[C],
H. Okumura[D], M. Miyahara[A]

KEK[A], AIST[B], RIKEN[C], Univ. of Tsukuba[D]

Pixel2024

November 20, 2024

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- Development of CIGS detector with high radiation tolerance
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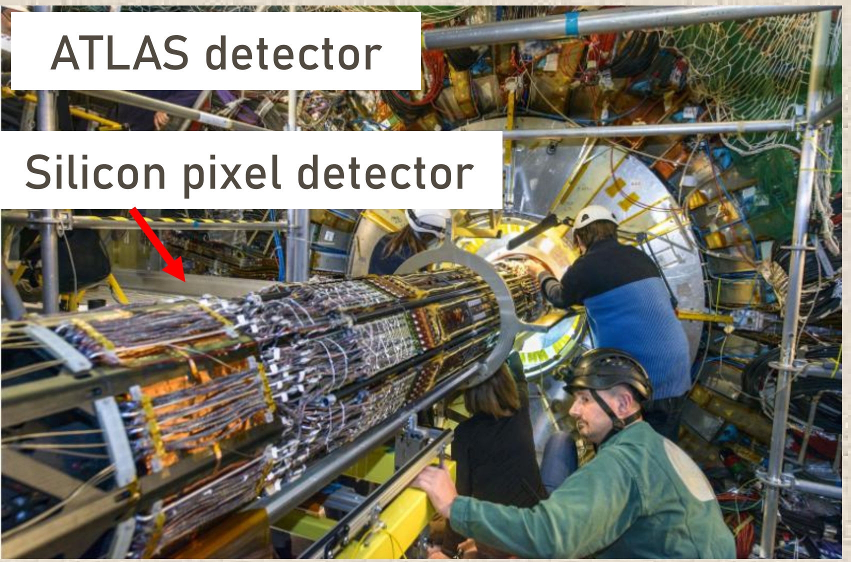
Particle detector in hadron collider experiment



In hadron collider experiments, silicon detectors are most widely used for particle tracking detectors.

The inner detector is exposed to high radiation levels, leading to the degradation of various semiconductor properties due to radiation damage.

Characters	LHC	HL-LHC	HE-LHC/FCC
Start year	On going	~2029	Future
Collison energy $\sqrt{s}$ [TeV]	14	14	27/100
Luminosity /LHC (0.7 MGy)	× 1	× 10	× 100
Particle detector	**Silicon**	**Silicon**	**??**

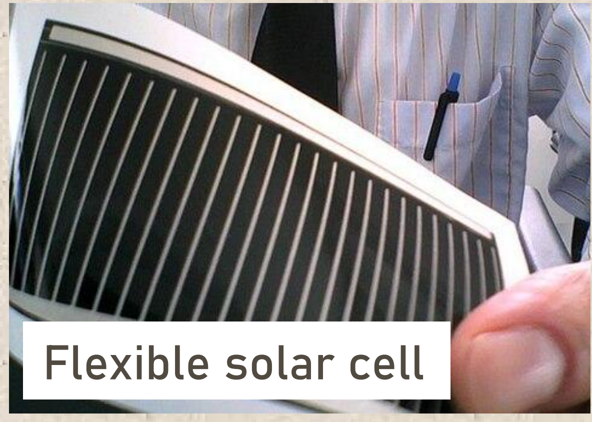
High energy experiment (LHC) plans to construct the higher energy and luminosity accelerator for the new particle search

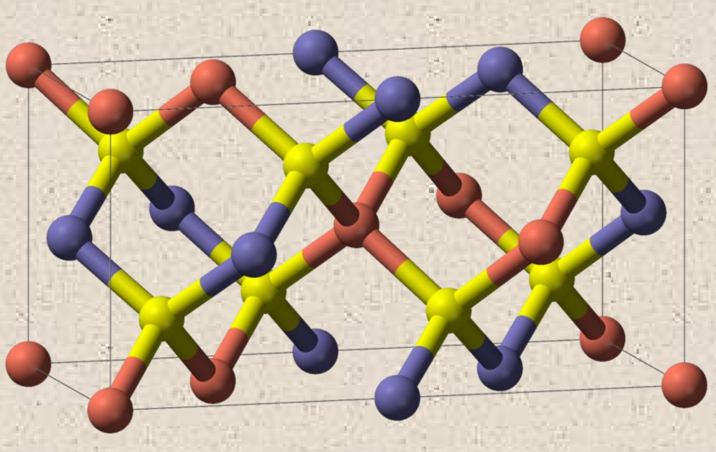
Need to development of high radiation tolerant particle detector
→ New semiconductor detector ??

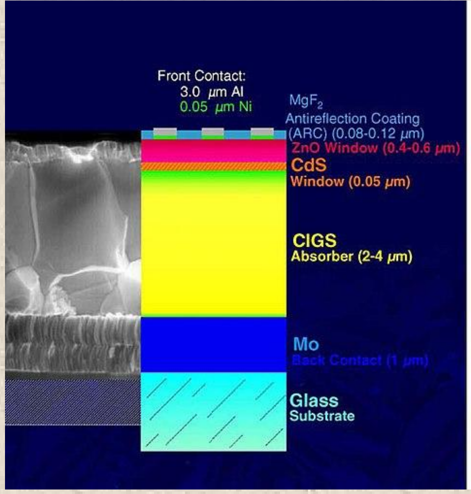
$\mathrm{Cu(In,Ga)Se_2}$ semiconductor (CIGS solar cells)

A CIGS is an alloy semiconductor of $\mathrm{CuInSe_2}$ and $\mathrm{CuGaSe_2}$, which is widely developed as a solar cell.

https://www.wikiwand.com/en/articles/Copper_indium_gallium_selenide_solar_cell


Flexible solar cell


CIGS unit cell. Red=Cu,Yellow=Se,Bule=In/Ga



CIGS has high absorption coefficient $> 10^5\ \mathrm{cm}^{-1}$ for photons with $\mathrm{E} \geq 1.5\ \mathrm{eV}$ $(\mathrm{Si}{\sim}10^3\mathrm{cm}^{-1})$

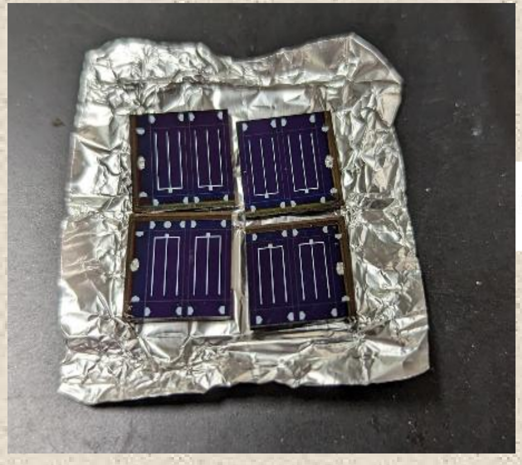
- High 20% efficiency (same level as Si) with 2-4 $\mu\mathrm{m}$ thick CIGS layer → Lightweight and Flexible
- Low deposition cost → possible to make large-area
- Radiation damage recovery with low temperature annealing $(\sim 100^\circ\mathrm{C})$

Radiation hardness semiconductor (CIGS)

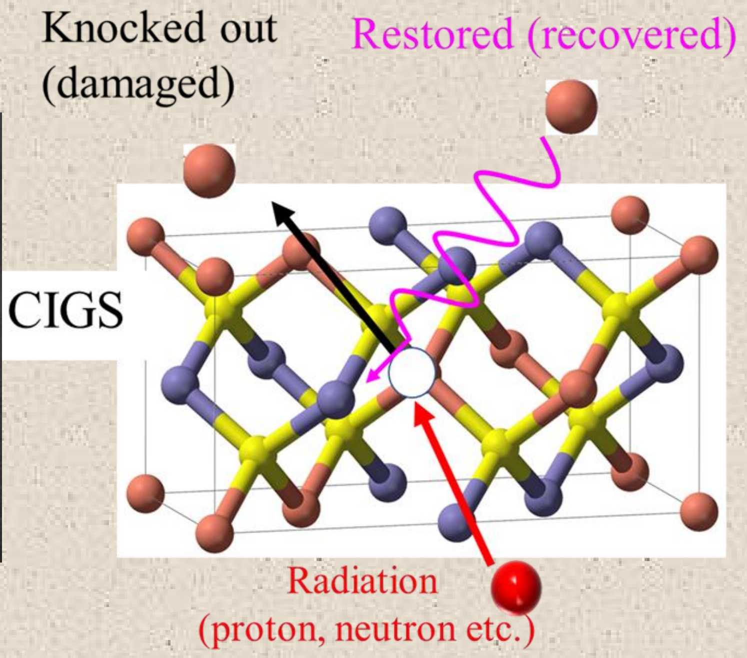
CIGS recovered radiation damage through heat annealing around $100°\mathrm{C}$

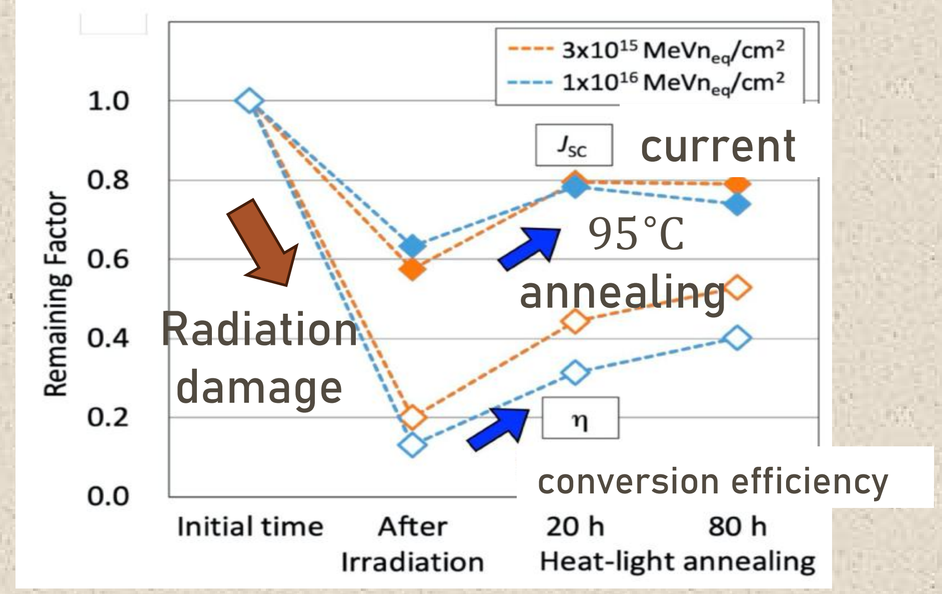
→ Suspended ions like Cu^+ expected to be restored to atomic defects

After irradiation
$10^{16}\ \mathrm{MeV} \cdot \mathrm{n_{eq}/cm^2}$



CIGS solar cells (AIST)





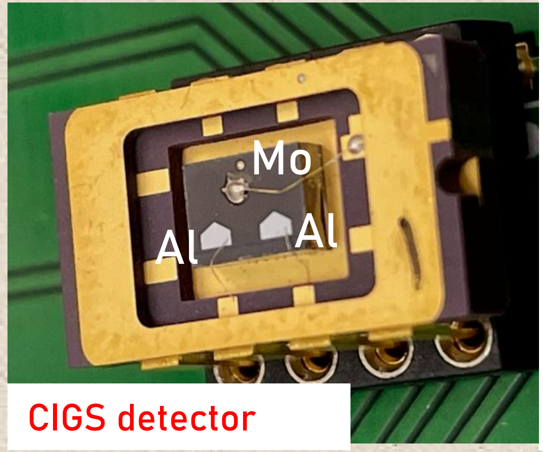
J. Nishinaga *et al.*, 2023 Jpn. J. Appl. Phys. 62 SK10

CIGS solar cells recovered from high radiation damage $(10^{16}\mathrm{MeV} \cdot \mathrm{n_{eq}/cm^2})$
→ CIGS semiconductor has the potential to be a particle detector with high radiation tolerance!

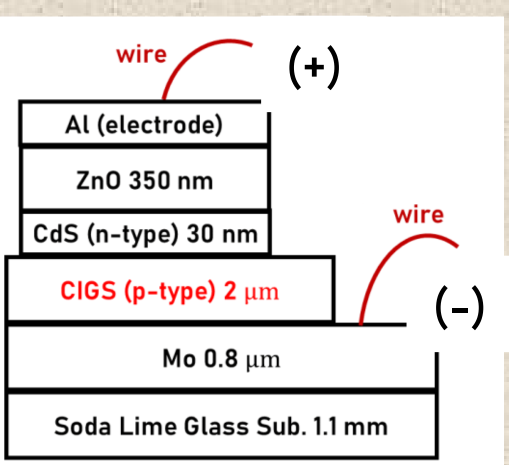
Development of CIGS detector

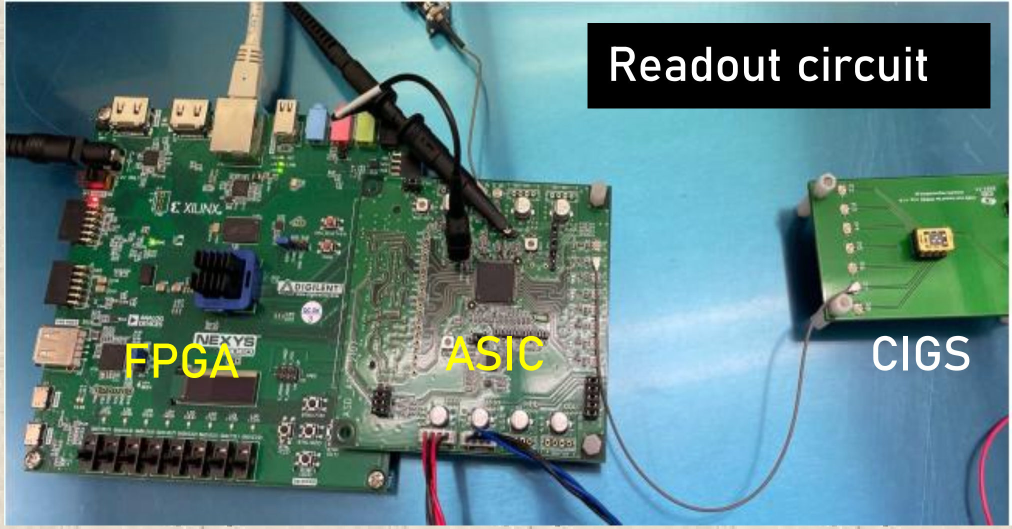
Specifications of CIGS detectors

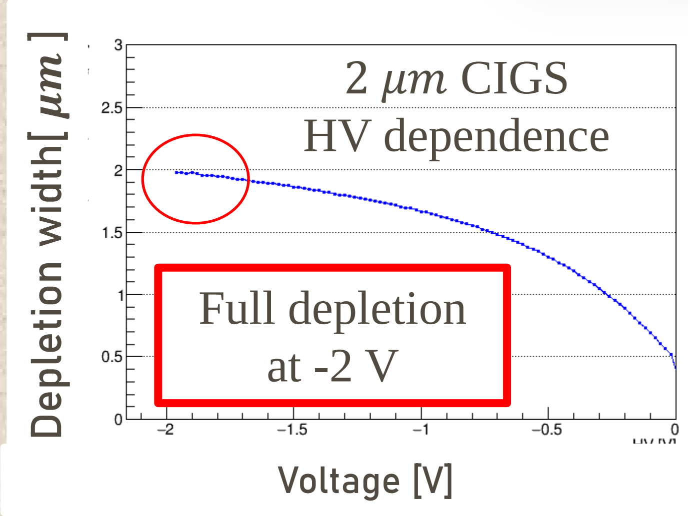
- p-type (CIGS), n-type (CdS)
- thickness $2\ \mu m$
- Active area : $5\ \mathrm{mm}^2/\mathrm{channel}$
- Operation Voltage : -2 V

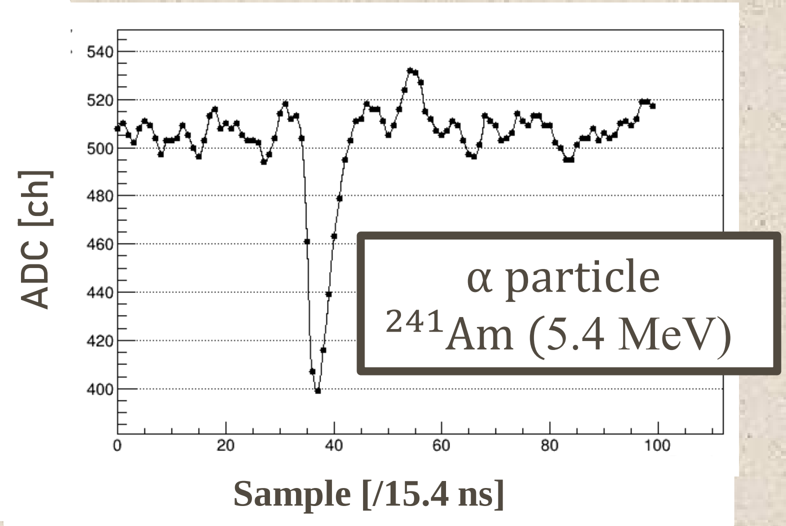


CIGS detector

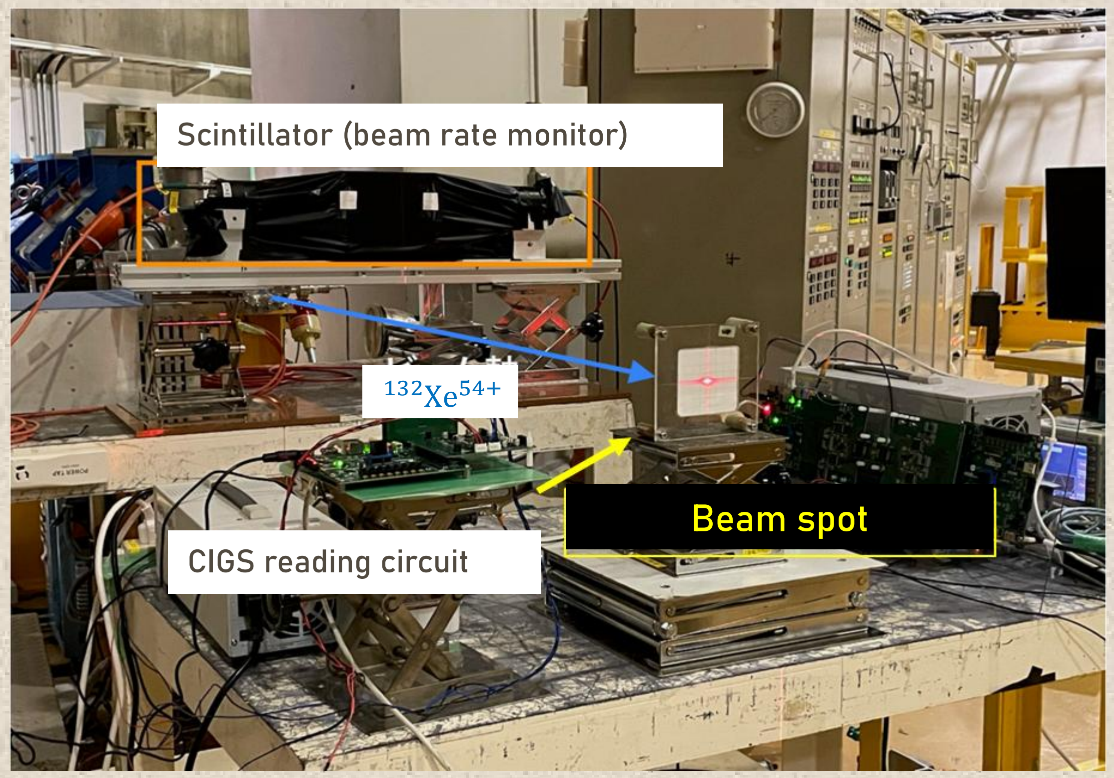

wire
(+)
Al (electrode)
ZnO 350 nm
CdS (n-type) 30 nm
wire
CIGS (p-type) 2 μm
(-)
Mo 0.8 μm
Soda Lime Glass Sub. 1.1 mm







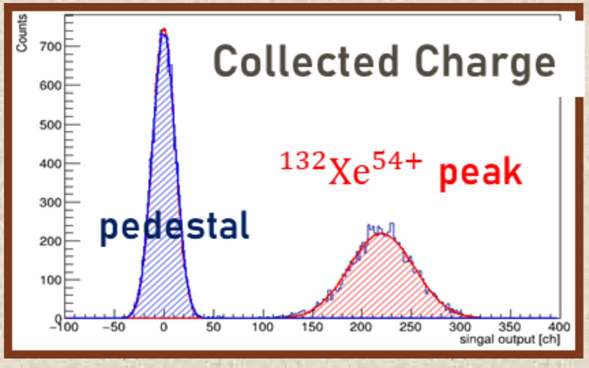
Irradiation experiment at HIMAC

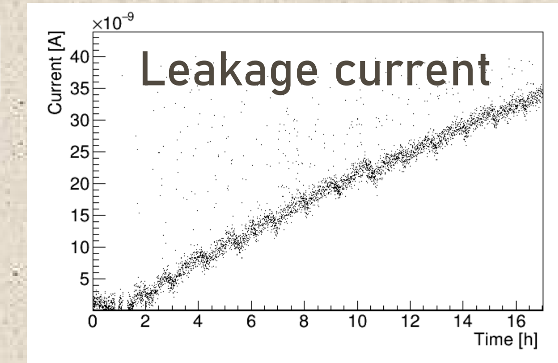


HIMAC Beam Condition

- Beam : $^{132}Xe^{54+}$ ion
- Energy : 400 (MeV/u)
- Beam size (ϕ) : 3-5 mm
- Fluence : 10^7 /3.3 s (ppp)

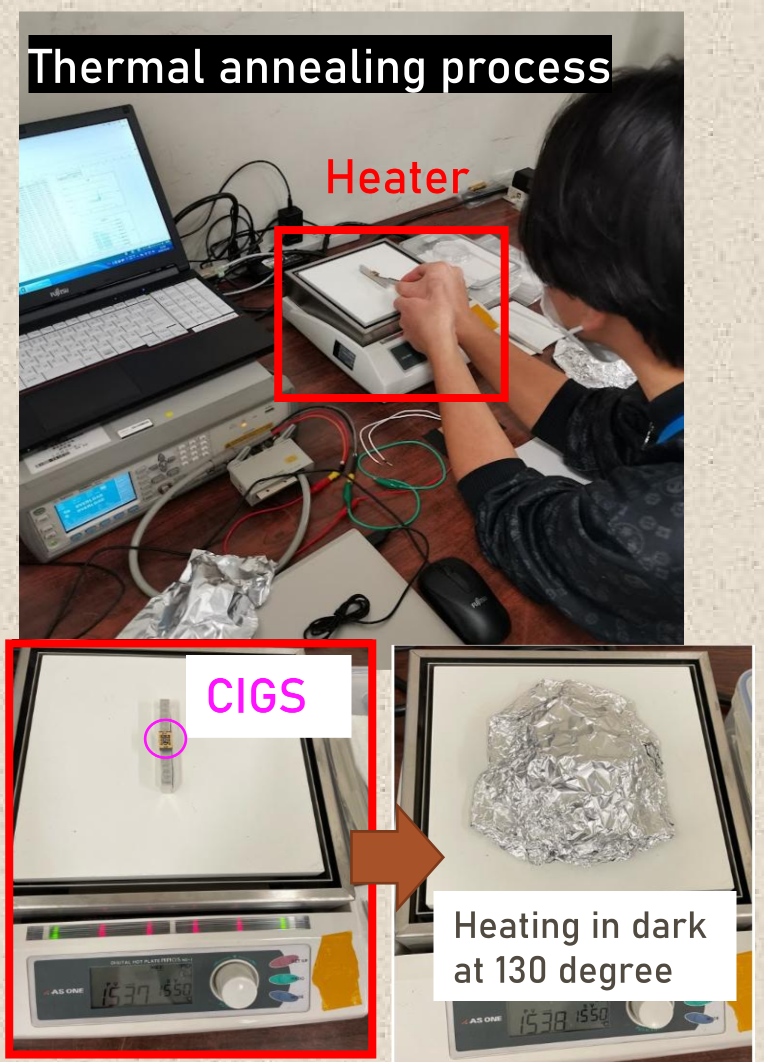
Measurement parameters



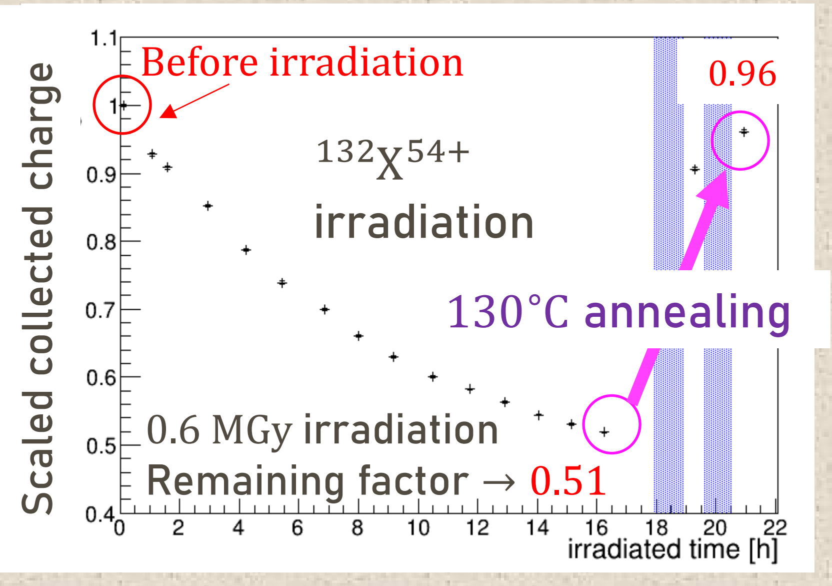


Investigation of detector performance recovery by thermal annealing
Detector performances : collected charge (Q) and leakage current (I)

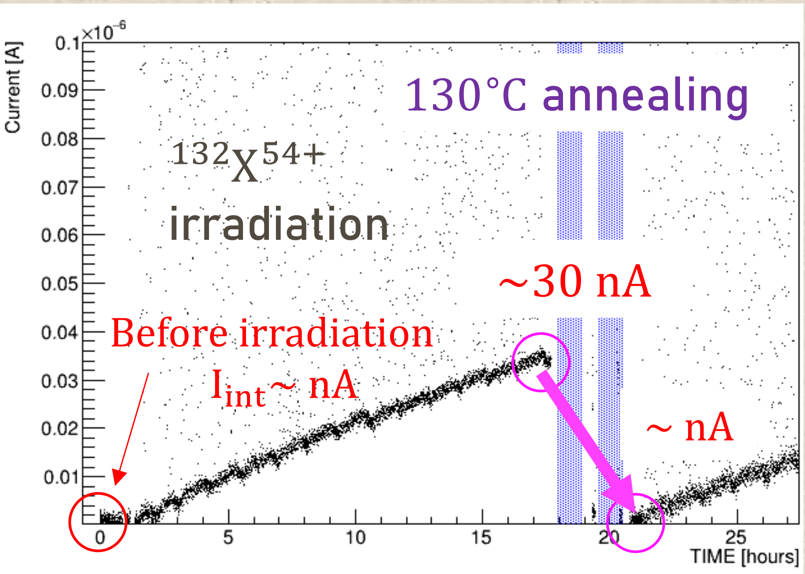
Study of the recovery mechanism by thermal annealing



Collected charge monitoring

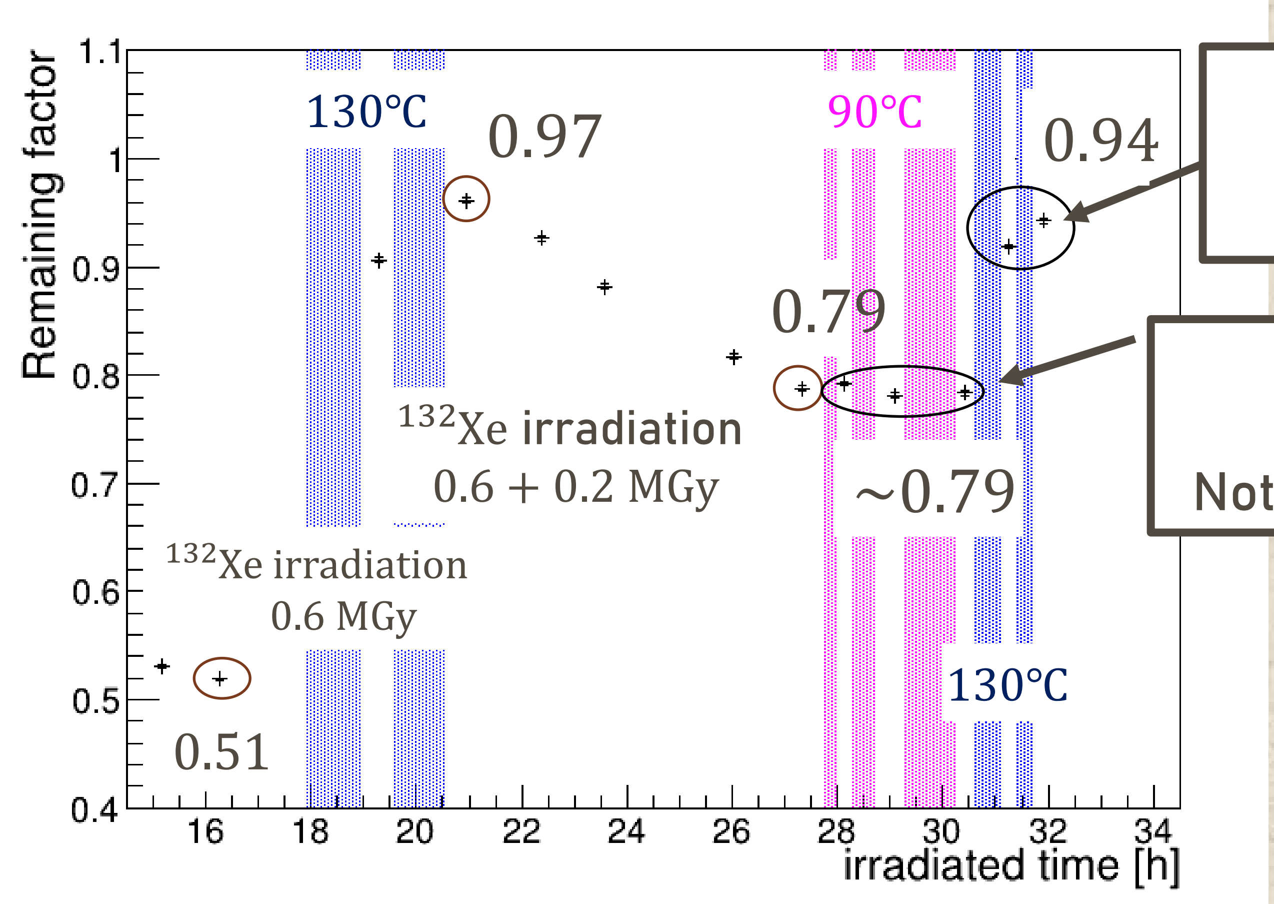


Leakage current monitoring



After 2 hours thermal annealing at 130 ℃,
Collected charge from $^{132}Xe^{54+}$ and leakage current
were recovered to the same level as before irradiation
→ Decreasing defect levels created by radiation damage

Study of the recovery system by thermal annealing (2)



M. Togawa et al., 2024 JINST 19 C05042

2nd 130℃ thermal annealing
(+30 min, +20 min)
0.79 → 0.94 (+50 min)

90℃ thermal annealing
(+15 min, +30 min, +1h)
Not sufficient recovery (insufficient annealing time)

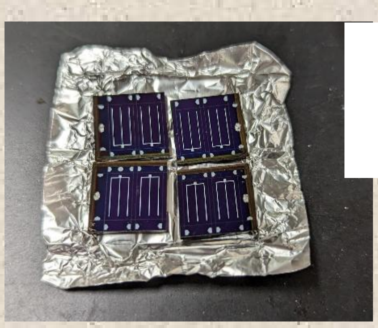
CIGS detector continuously recovered through short-term annealing at 130℃

→ **Confirmed to have a high radiation tolerance as a particle detector!**

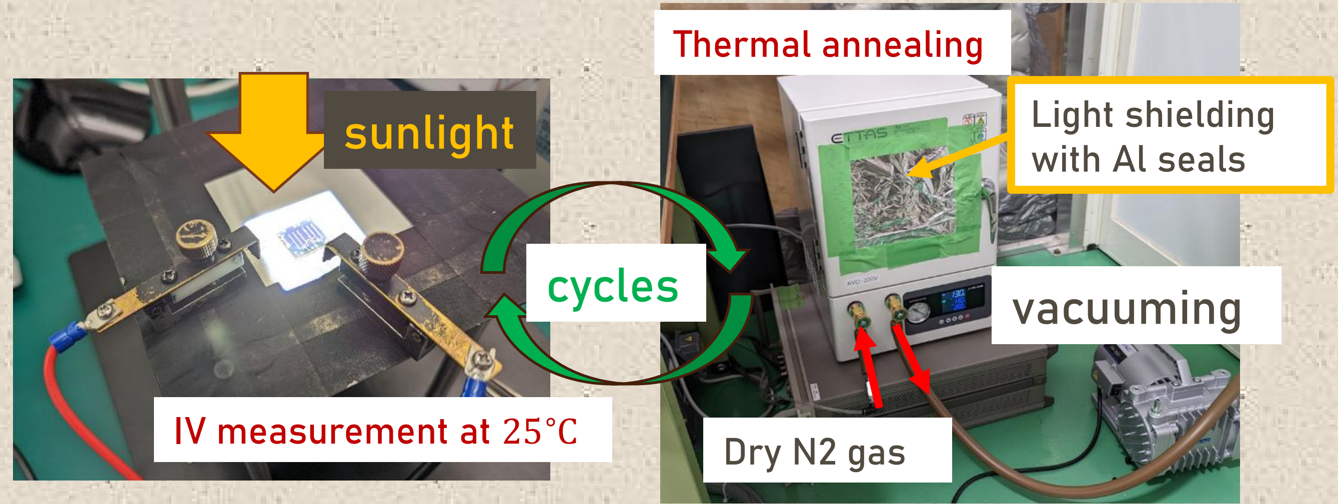
Recovery dependency on annealing condition

Irradiated 70 MeV proton to CIGS solar cells $(7 \times 10^{15}\ \mathrm{n_{eq}})$ at CYRIC

- Measured current value $(\mathrm{J_{SC}})$ with zero bias voltage at $25^\circ\mathrm{C}$
- Three difference annealing process $(90^\circ\mathrm{C}, 110^\circ\mathrm{C}, 130^\circ\mathrm{C})$

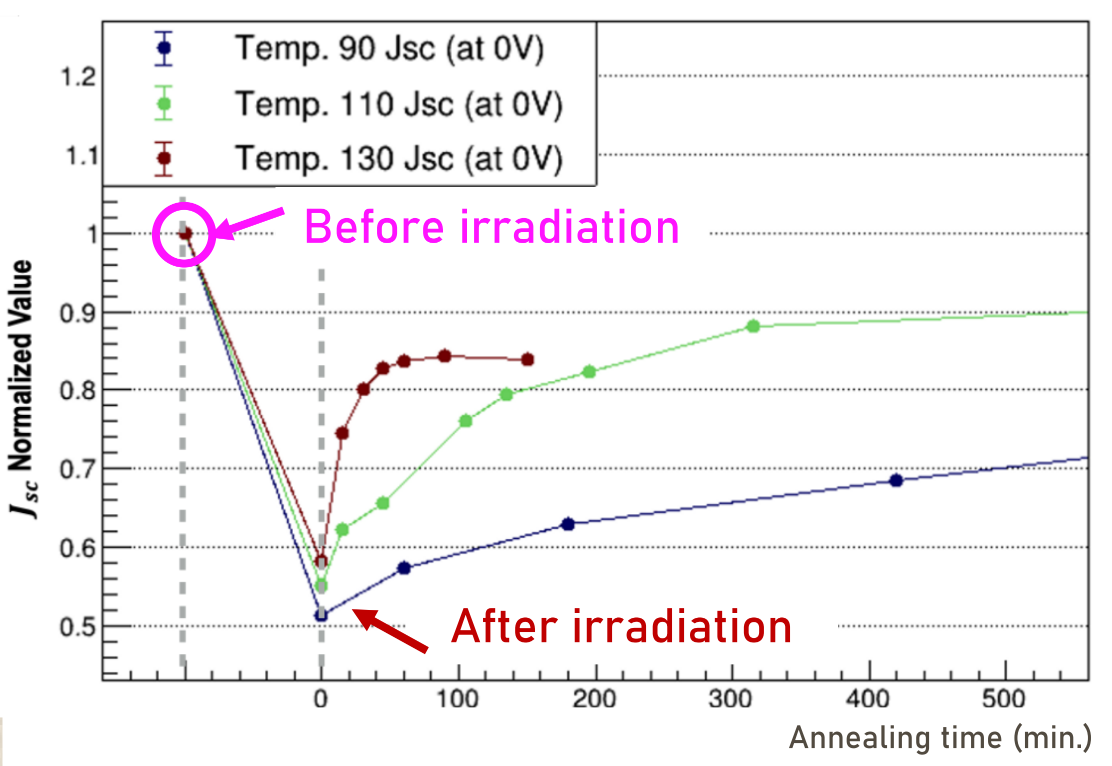


CIGS solar cells after proton irradiation



The recovery speed of current under sunlight shows strong temperature dependence between $90^\circ\mathrm{C}$ and $130^\circ\mathrm{C}$.

K. Itabashi et al., NIMA A, 1067 (2024) 169637.

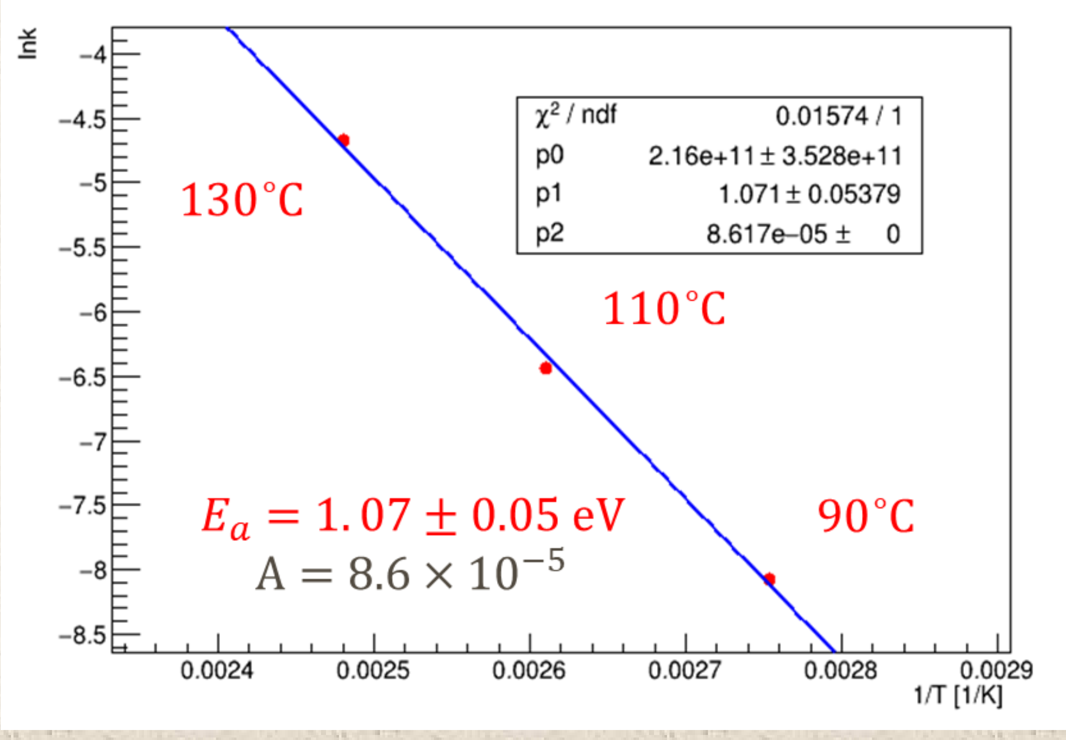


Excitation energy during thermal annealing

Arrhenius plot : $\ln(k) = -\frac{E_a}{k_b T} + \ln(A)$ $(\mathrm{k}: \text{Reaction rate constant})$

(A : constant value, k_b: Boltzman const.)

Arrhenius plot with CIGS soler cells



$E_a = 1.07 \pm 0.05$ eV
$\mathrm{A} = 8.6 \times 10^{-5}$

[1] A. Zunger et al., 26th IEEE Phytovoltaic Specialists Specialists Conf. (1997).

defect	Creation energy [eV] [1]
V_{Cu}^{-}	0.63
V_{In}^{-}	3.21
V_{In}^{2-}	3.62
V_{In}^{3-}	4.29
Cu_{In}^{-}	1.83
Cu_{In}^{2-}	2.41
In_{Cu}^{+}	1.85
In_{Cu}^{2+}	2.55
In_{Cu}^{3+}	3.34

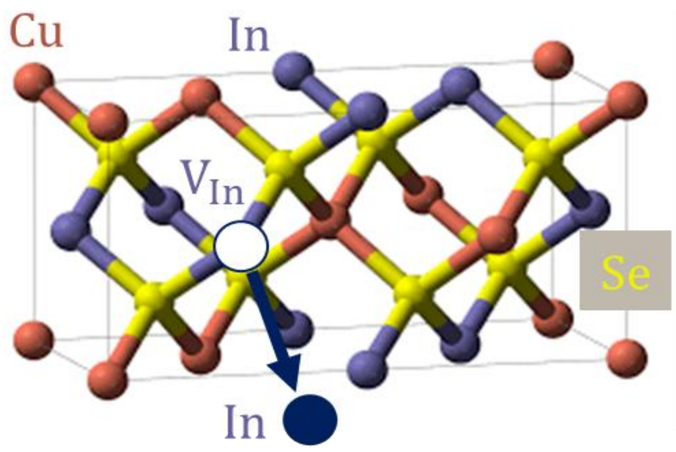
Excitation energy $E_a \sim 1.0$ eV during thermal annealing between $90 - 130$°C

Cupper ions are activated and created cupper vacancies
$\rightarrow Cu^{+} + V_{Cu}^{-}$

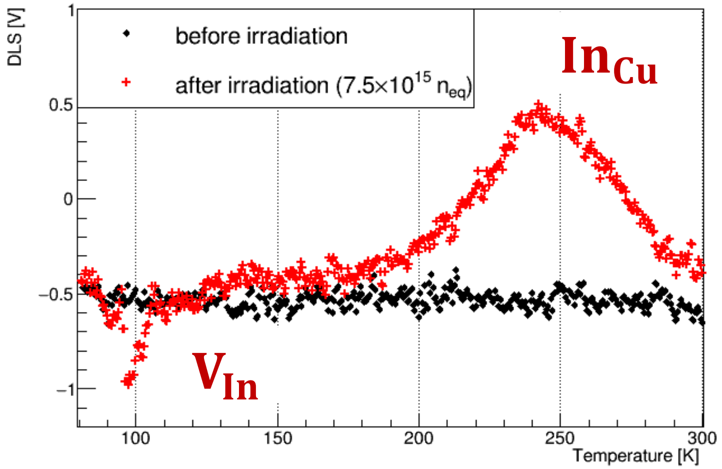
Defects measurement by DLTS after proton irradiation

Deep Level Transient Spectroscopy (DLTS) is major technique used to measure deep-level defects in semiconductor.

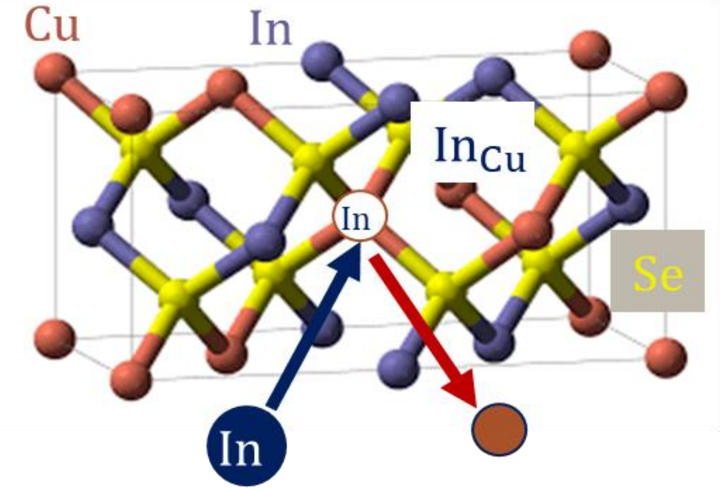
V_{In} defects



DLTS signals



$\mathrm{In_{Cu}}$ defects

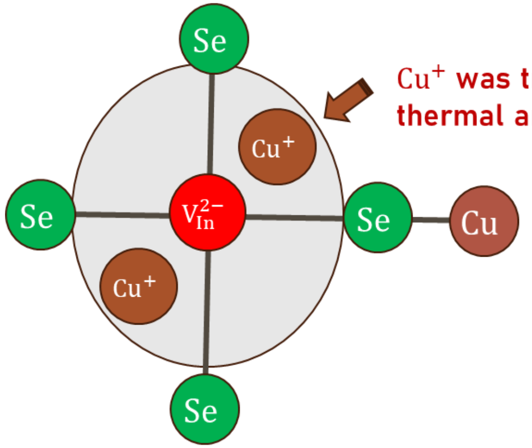


After proton irradiation with $7 \times 10^{15}\ \mathrm{MeV} \cdot \mathrm{n_{eq}/cm^2}$, In-vacancy ($\mathrm{V_{In}}$) and Cu-site-In ($\mathrm{In_{Cu}}$) defects are observed by DLTS measurement.

Defects measurement by DLTS after Annealing

After 130℃ annealing for one hour, radiation defects (V_{In}, In_{Cu}) are decreased.

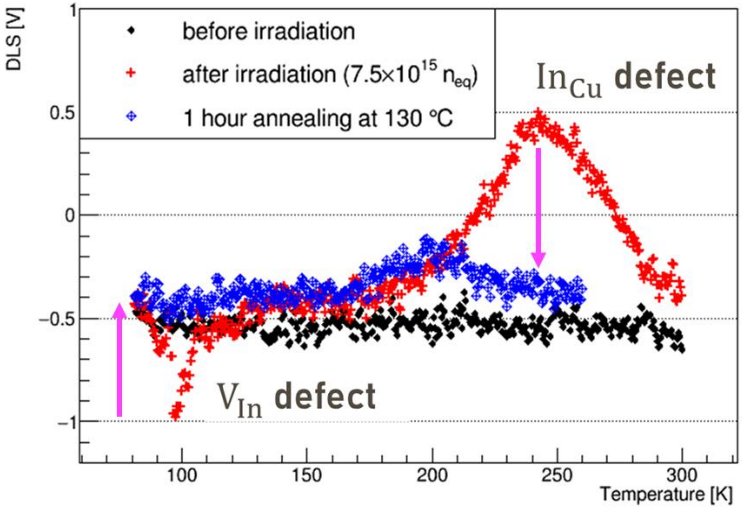
V_{In} defects



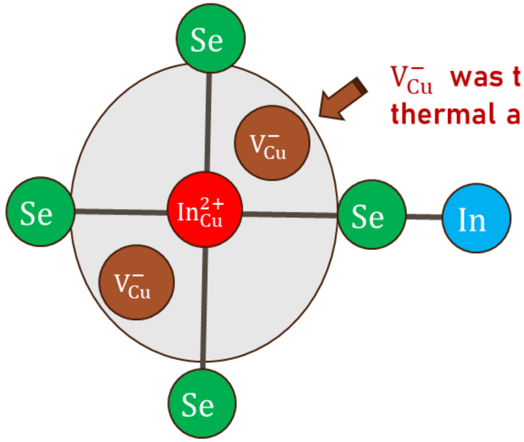
Cu^{+} was trapped by thermal annealing

Electrical neutrality($V_{In}^{2-} + 2Cu^{+}$)

DLTS signals



In_{Cu} defects



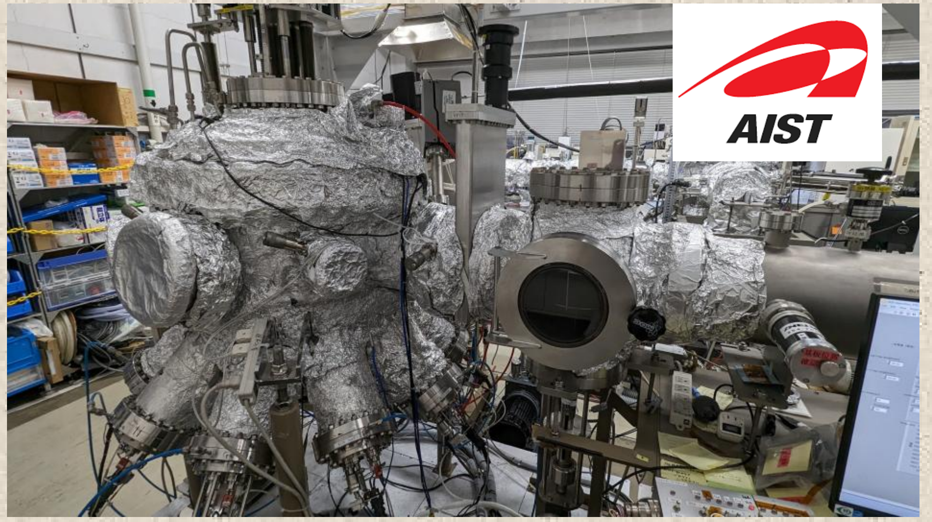
V_{Cu}^{-} was trapped by thermal annealing

Electrical neutrality($In_{Cu}^{2+} + 2V_{Cu}^{-}$)

As a result of Arrhenius plot fitting, Cu^{+} and V_{Cu}^{-} ions are activated during annealing.

→ V_{In} and In_{Cu} defect levels are reduced by neutralizing with Cu^{+} and V_{Cu}^{-} ions

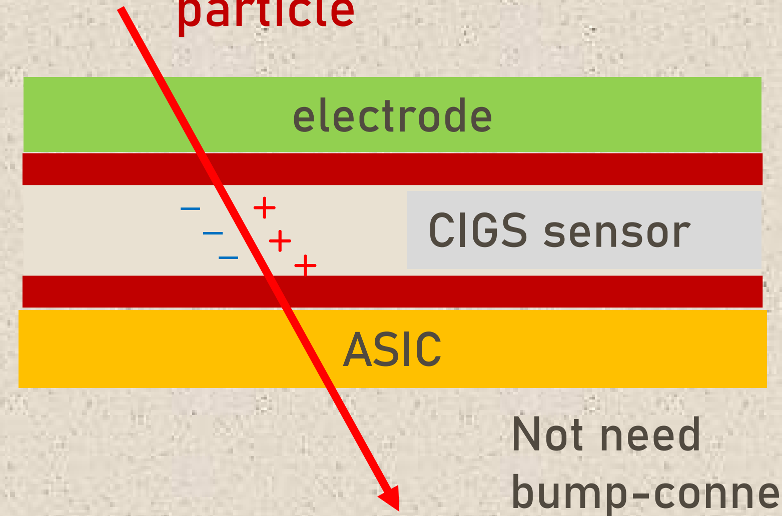
Outlook for CIGS detector development



CIGS deposition process :

- Low-temperature $300 \sim 500°\mathrm{C}$
- High deposition rate $\sim 1\ \mu\mathrm{m/hour}$
- Direct deposition onto the substrate

Due to the low thermal load on the substrate, direct deposition onto ASICs is promising.



Not need
bump-connection

We aim to develop a pixel-type CIGS detector by directly depositing CIGS onto an ASIC substrate.

Conclusions and outlook

Conclusions

- The CIGS semiconductor is known to recover from radiation damage, and this ability also actives for the particle detector confirmed by the HIMAC experiment.
- As a result of DLTS measurement, the defect levels of $\mathrm{V_{In}}$ and $\mathrm{In_{Cu}}$ were observed in the temperature range from $80\mathrm{K}$ to $300\ \mathrm{K}$ after proton irradiation with a fluence of $7 \times 10^{15}\ \mathrm{MeV} \cdot \mathrm{n_{eq}/cm^2}$. Moreover, these defects were recovered by $130°\mathrm{C}$ annealing for 1 hour.
- From Arrhenius plot fitting, the $\mathrm{V_{Cu}^-}$ and $\mathrm{Cu^+}$ were activated during thermal annealing ($\rightarrow \mathrm{Cu^+} + \mathrm{V_{Cu}^-}$) and they expected to bind $\mathrm{V_{In}}$ and $\mathrm{In_{Cu}}$ defects to achieve electrical neutrality.

outlook

- CIGS adopts a direct deposition method onto the substrate, and due to the low deposition temperature, it exerts minimal thermal stress on the substrate. We are currently attempting direct deposition onto the ASIC.

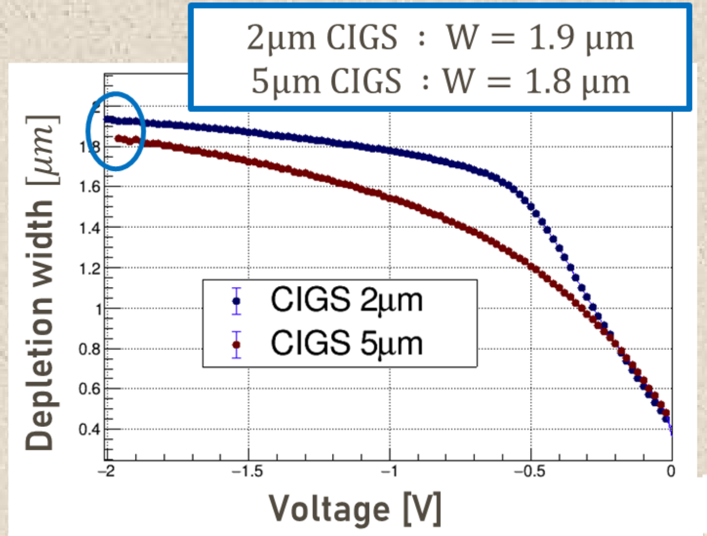
Thank you for your attention!

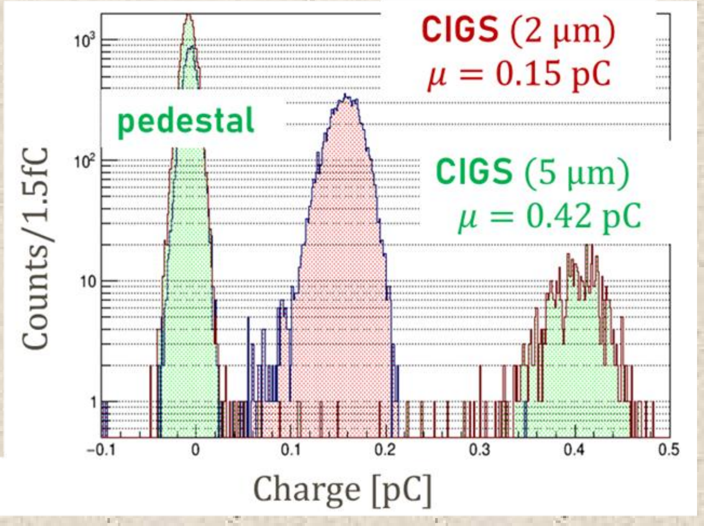
Backup

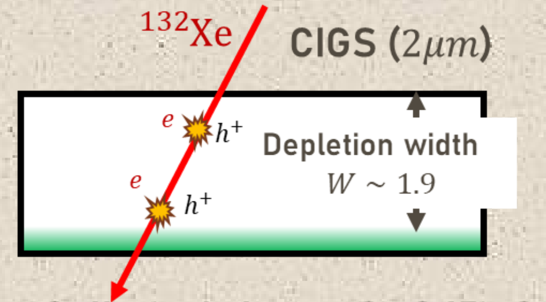
Thickness dependence of CIGS detector peformances

The collected charge is proportional to depletion width ($Q \propto W$).

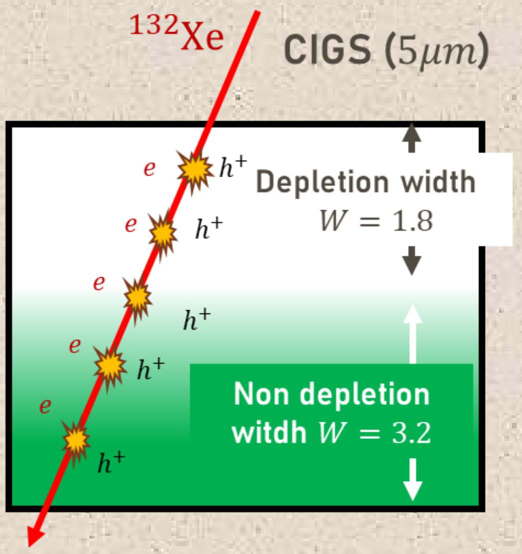
Both of depletion width (V=-2V) are about $2\ \mu\mathrm{m}$, but collected charge of $5\ \mu\mathrm{m}$ CIGS detector was **2.5 times larger** than one of $2\ \mu\mathrm{m}$ CIGS detector


2μm CIGS : $W = 1.9\ \mu\mathrm{m}$
5μm CIGS : $W = 1.8\ \mu\mathrm{m}$





Collected charge Q= 0.15 pC



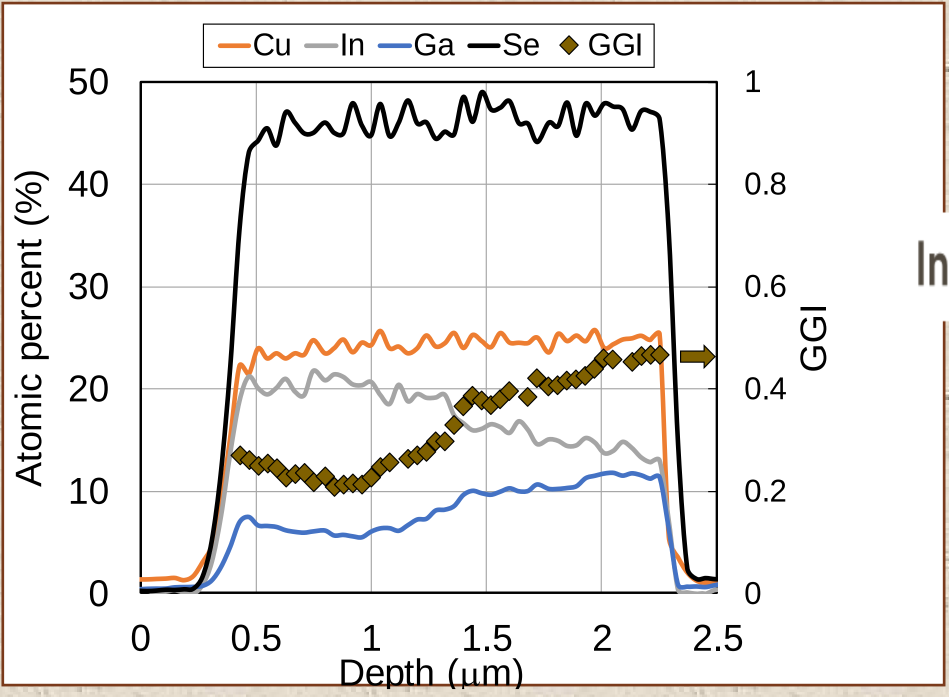
Collected charge Q= 0.42 pC

Is it possible to collect charges in non-depletion region ??

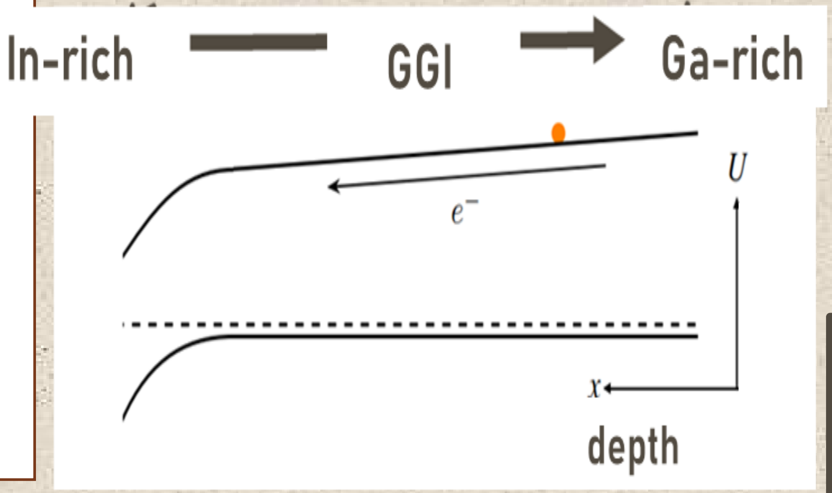
Energy gradient of CIGS layer

CIGS is an alloy semiconductor $\mathrm{CuInSe_2}$ and $\mathrm{CuGaSe_2}$.
Energy gap of CIGS changes with Ga composition ratio (GGI=[Ga]/[In]+[Ga]).

$$1.01\ \mathrm{eV}\ [\mathrm{GGI}=0] < \mathrm{E_g} < 1.64\ \mathrm{eV}\ [\mathrm{GGI}=1]$$



CIGS layer has GGI gradient for depth direction
→ **Promoting electron diffusion**



Not need for full depletion

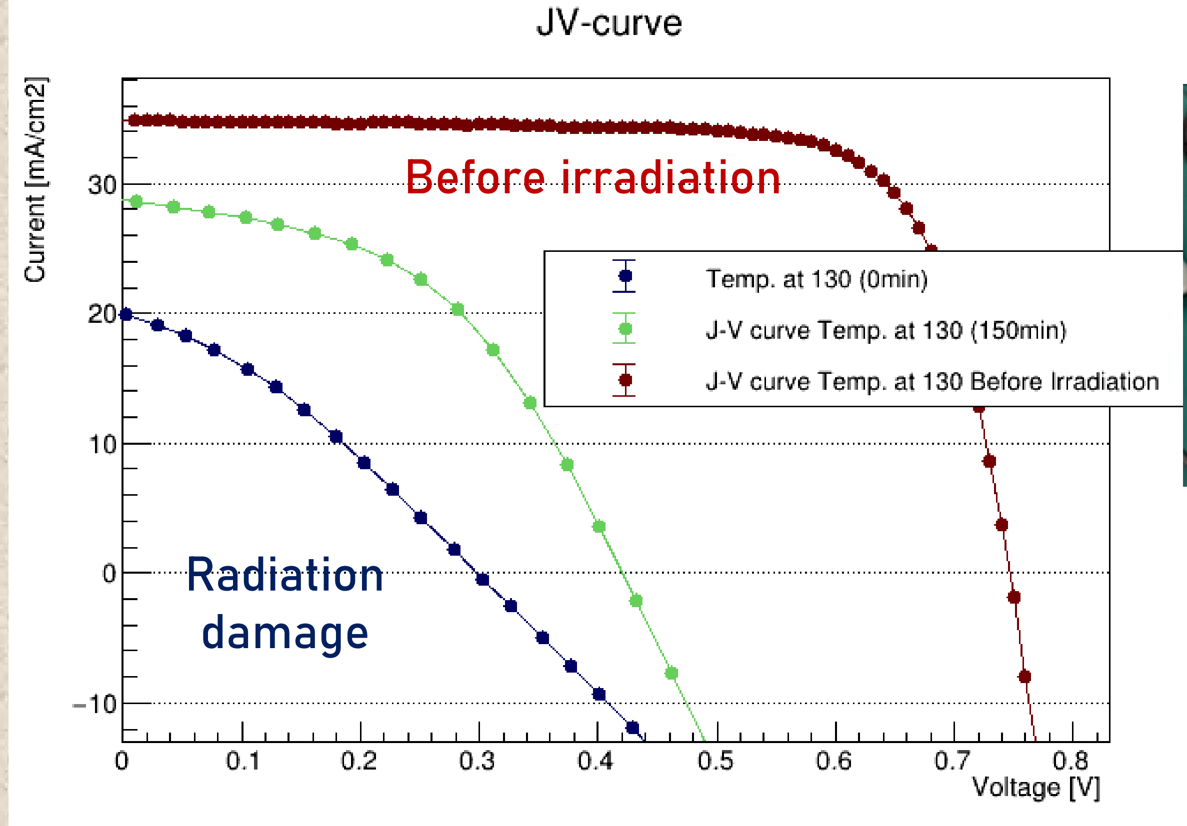
It allows
low voltage operation!

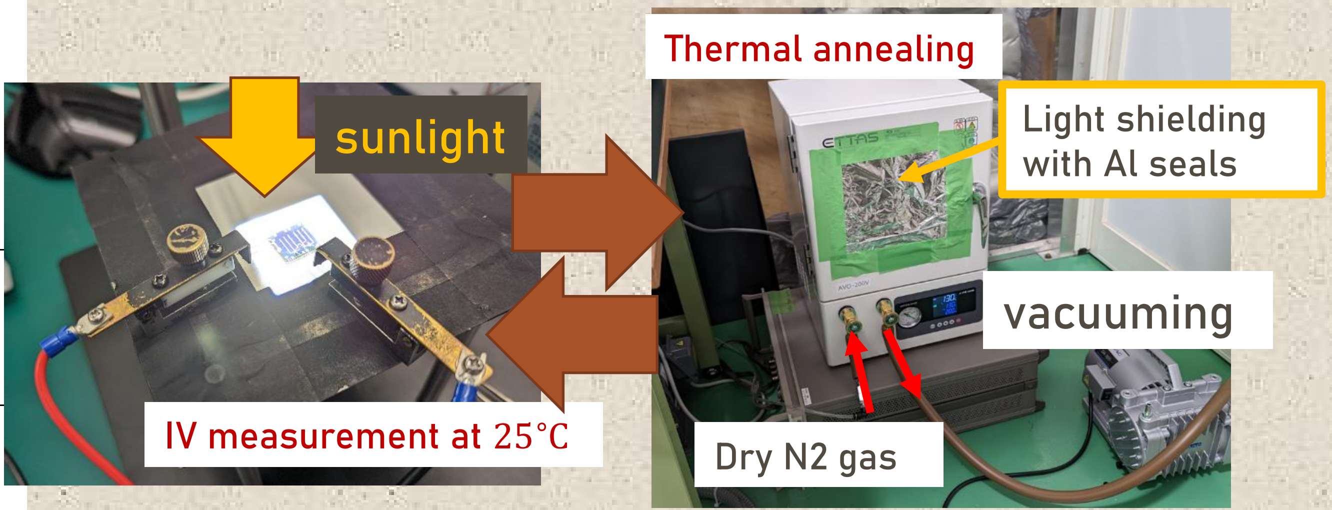
Proton irradiation experiment at CYRIC

CIGS solar cells (AIST)

CYRIC experiment : Irradiated 70 MeV proton to CIGS solar cells ($7 \times 10^{15}\ \mathrm{n_{eq}}$)

→ Study the heating time and temperature dependences of recovery mechanism





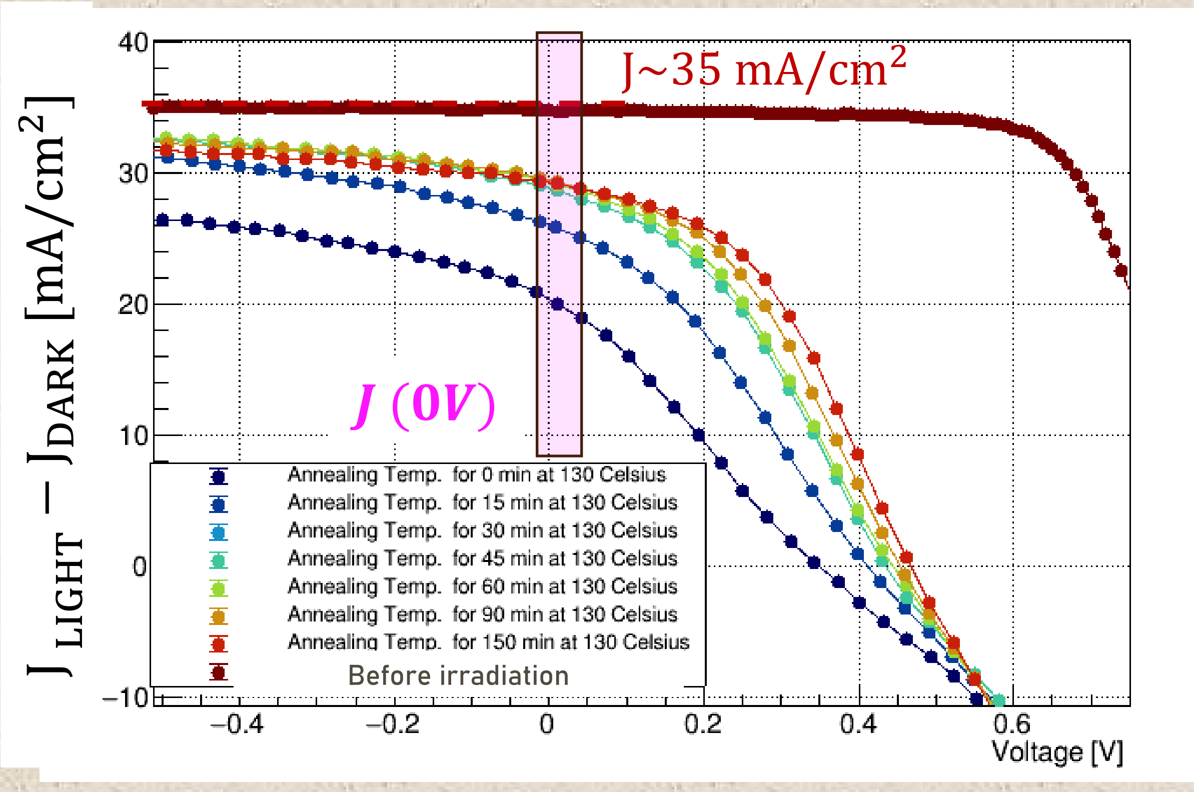
IV curve of CIGS solar cells

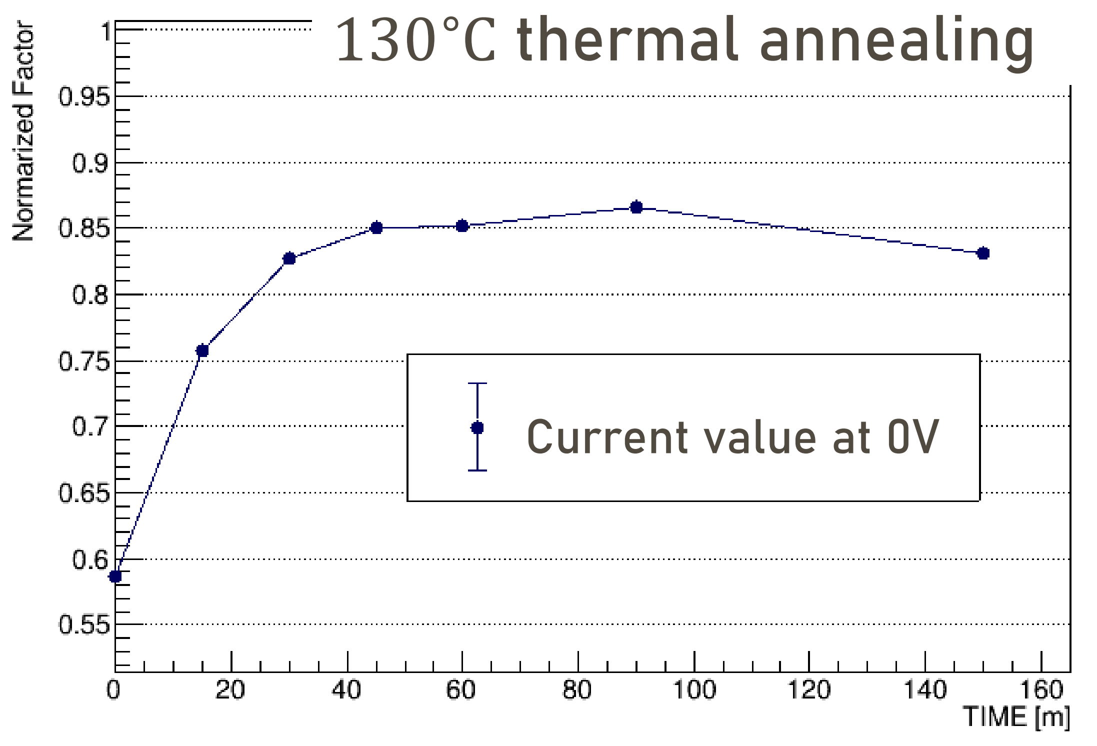
- J $[\mathrm{mA/cm^2}]$ decreased due to proton irradiation
  $\mathrm{J_{sc}(0V)} : 35 \rightarrow 20\ \mathrm{mA/cm^2}$
- Recovered J by thermal annealing (2.5 h) at 130°C
  $\mathrm{J_{sc}(0V)} : 20 \rightarrow 28\ \mathrm{mA/cm^2}$

Current recovery dependence of annealing time

The measurement current with incident sunlight is including dark current.

Excluded dark current : $J = \mathrm{J_{LIGHT}} - \mathrm{J_{DARK}}$

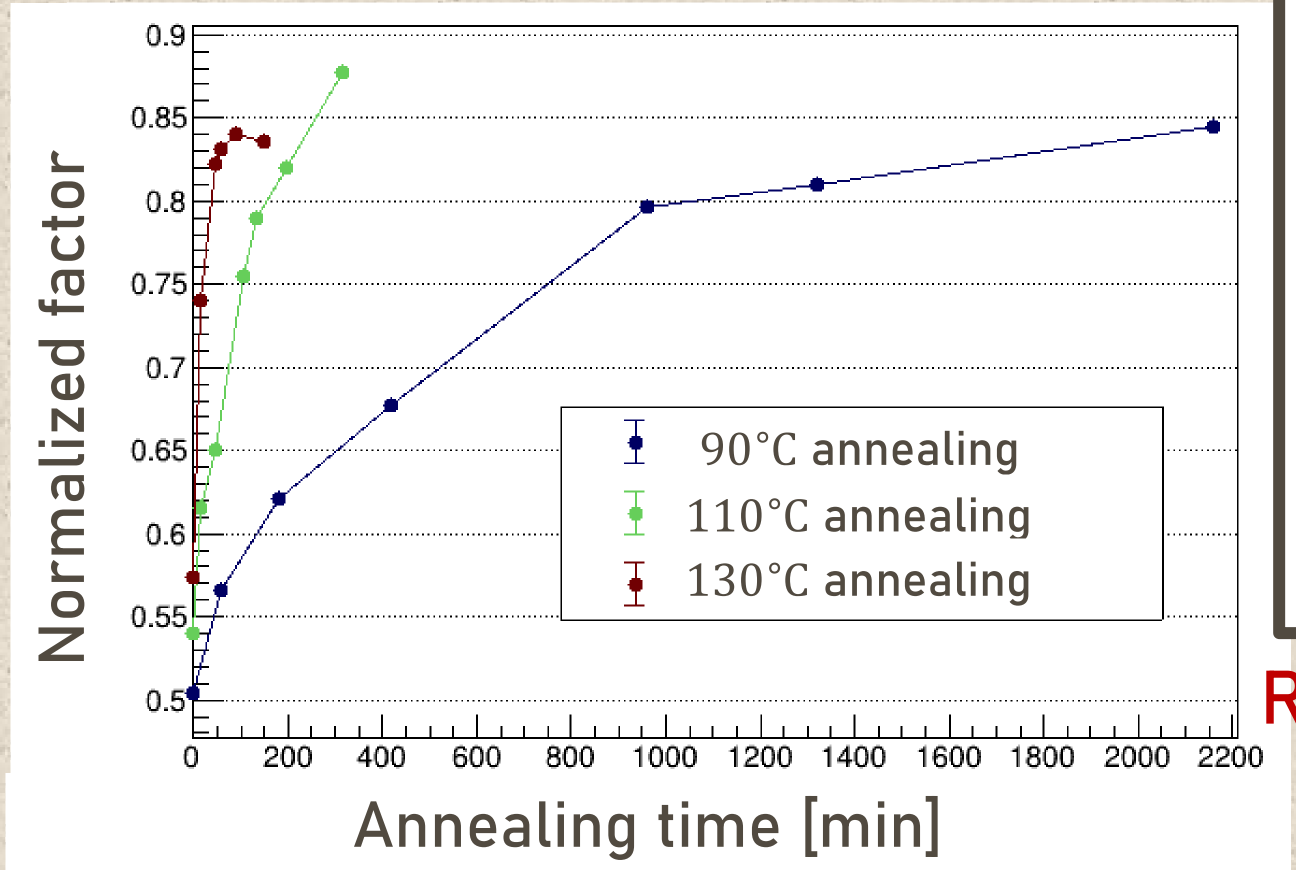




J recovered rapidly to saturation value (~1h)

Heating temperature dependence of recovery speed

I annealed CIGS solar cells at three differential temperatures (90°C, 110°C, 130°C).



130°C annealing : $J_{0V} = 0.57 \rightarrow 0.85\ (1h)$

90°C annealing : $J_{0V} = 0.50 \rightarrow 0.57\ (1h)$

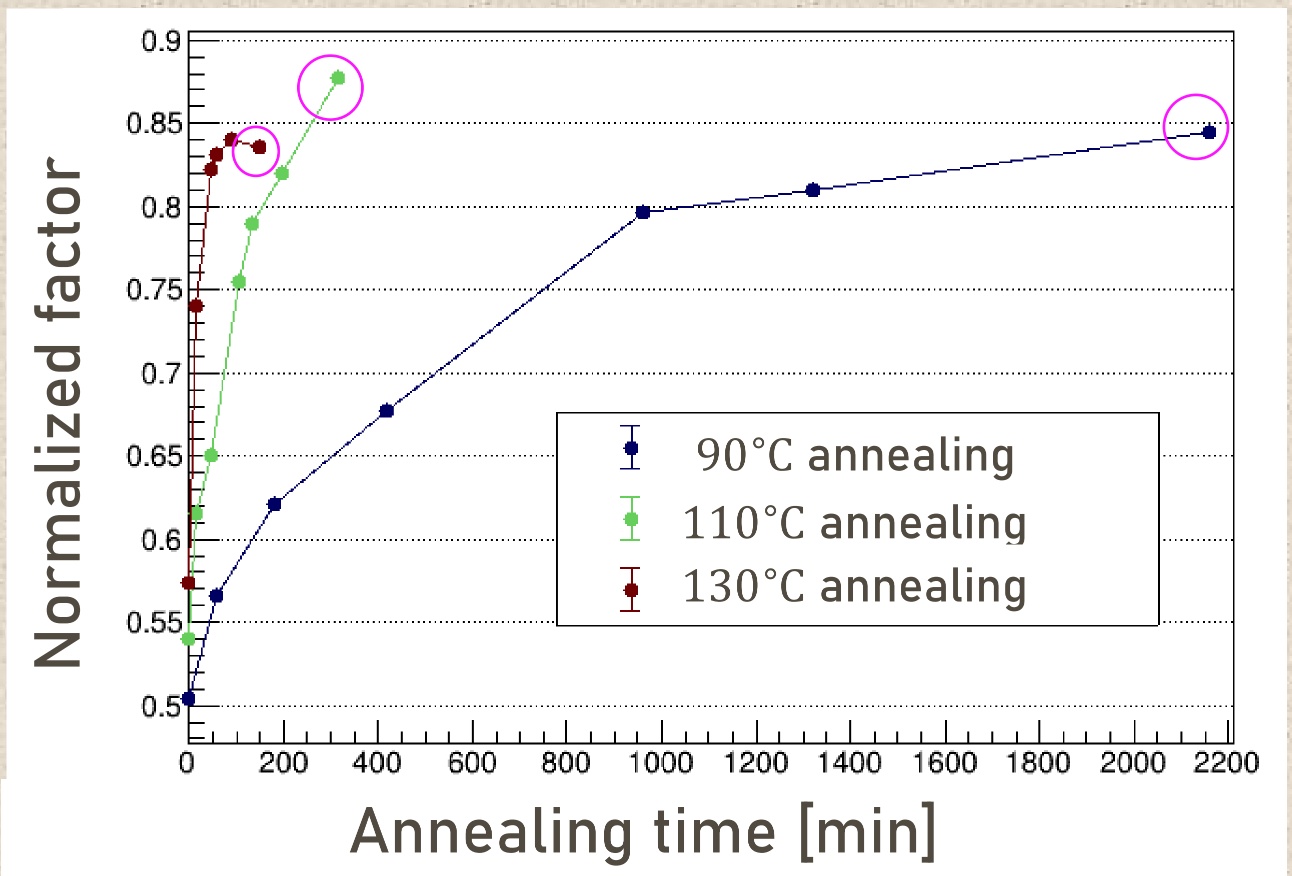
Comparable with recovery speed of collected charge at HIMAC experiment

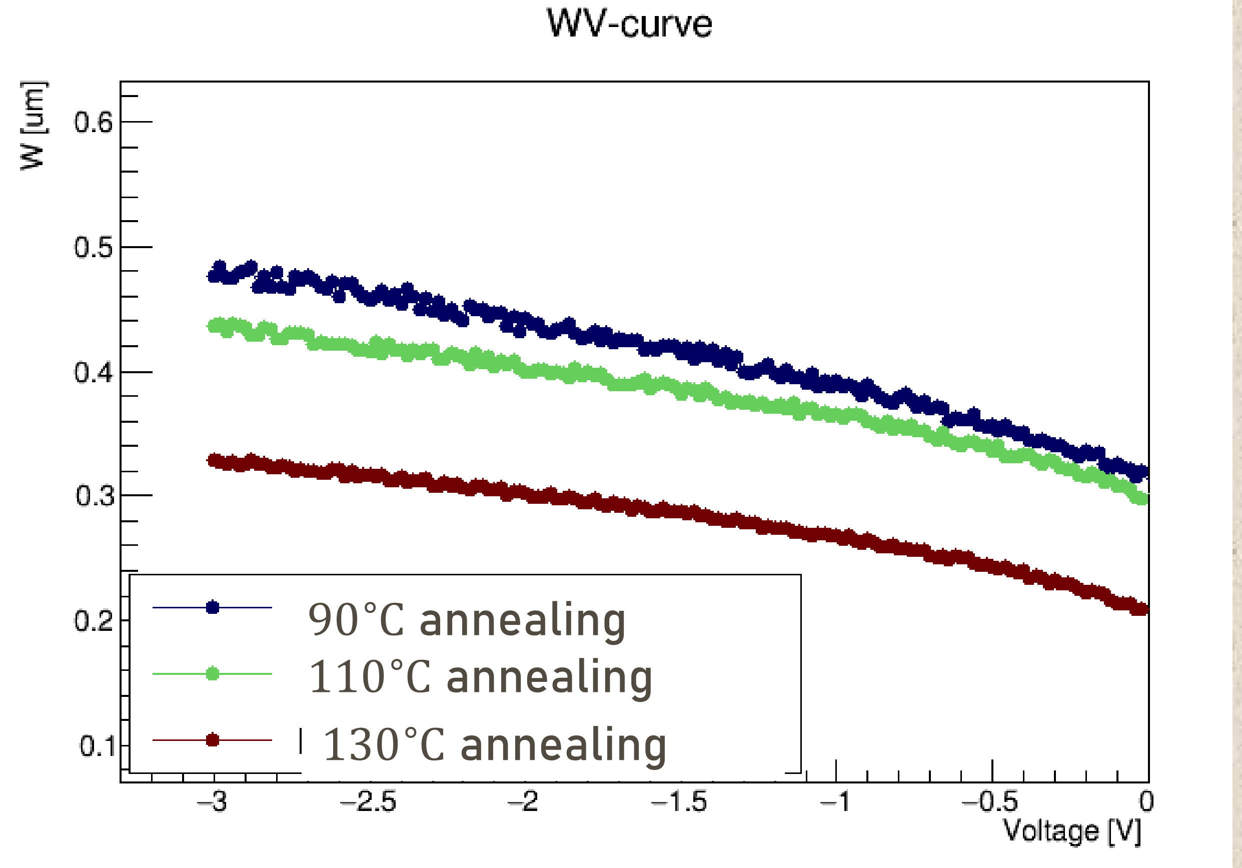
130°C annealing: $Q = 0.79 \quad \rightarrow 0.94\ \ (1h)$

90°C annealing: $Q = 0.79 \quad \rightarrow 0.79\ (1h)$

Recovery speed is greatly depending on heating temperature

Depletion width at each annealing time





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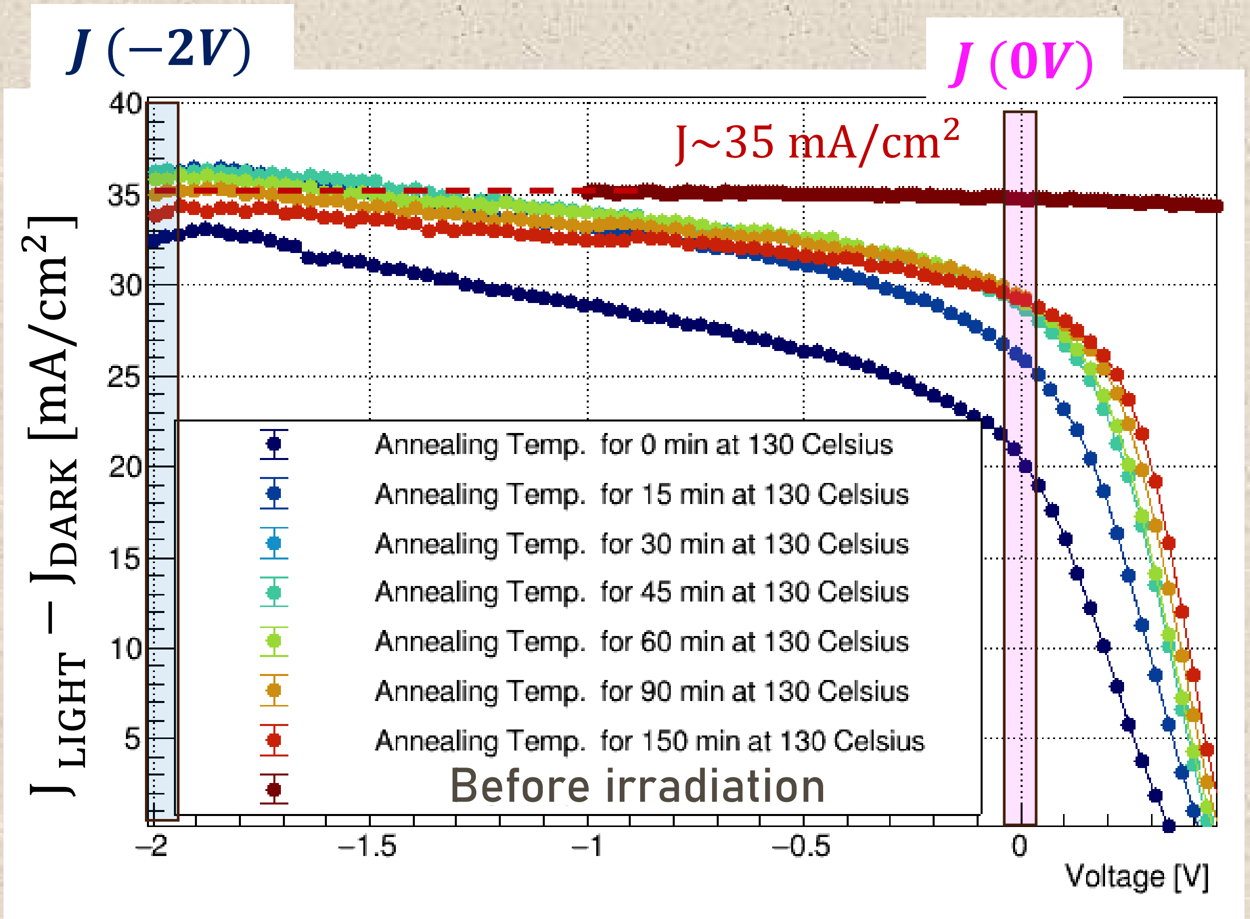
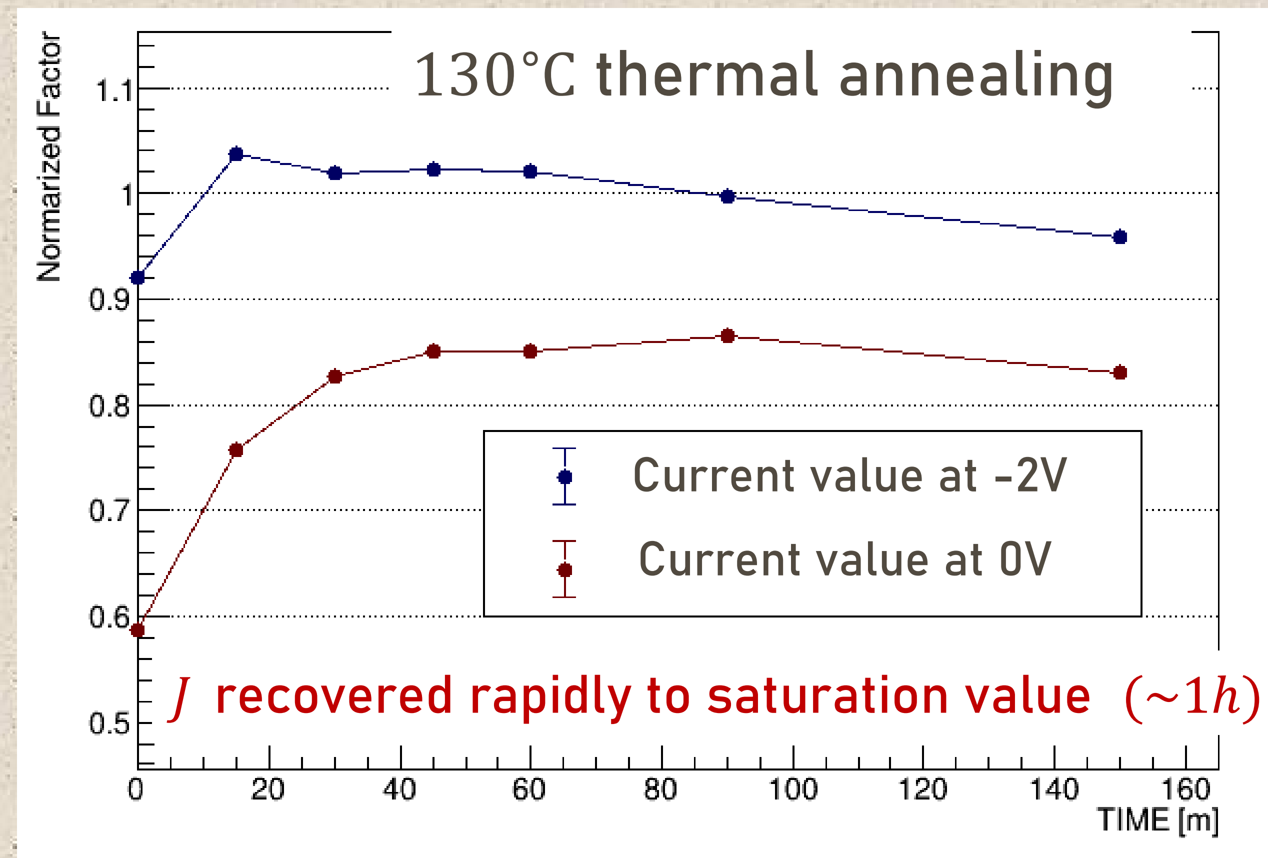




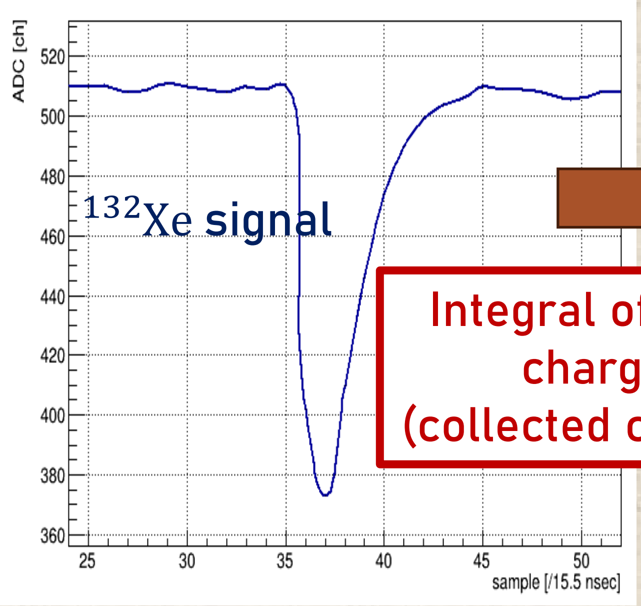
J recovered rapidly to saturation value $(\sim 1h)$

1. Study of Thickness dependence of CIGS (Collected Charge from xenon signal)

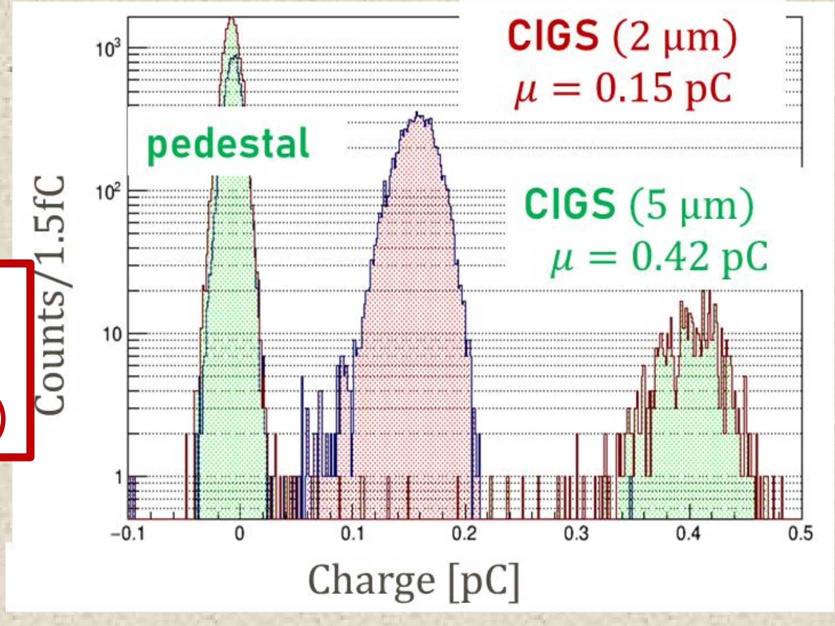
Collected charge evaluation: Xe beams (p=400 MeV/u) were irradiated to 2 µm and 5 µm CIGS semiconductor detectors, respectively.

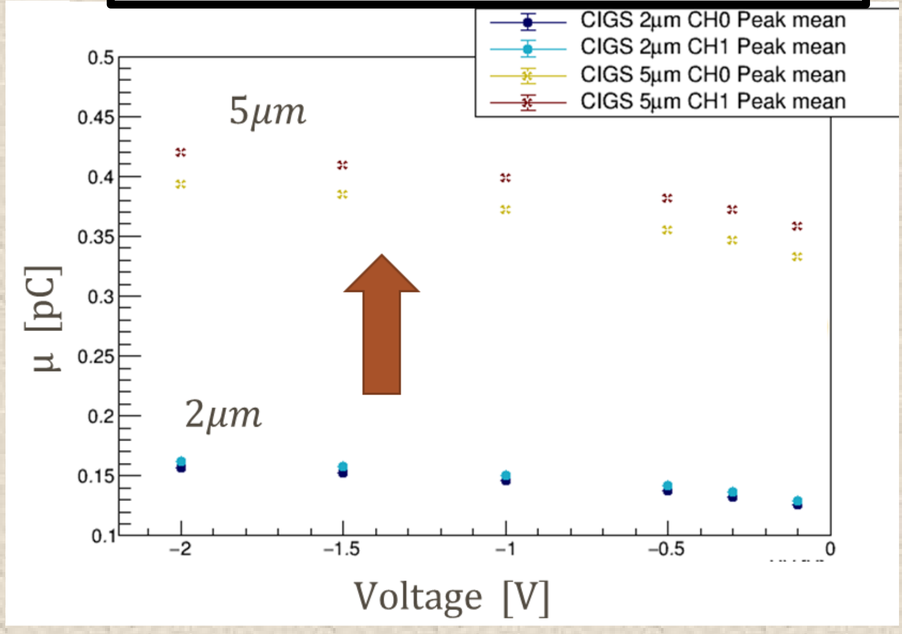
$Q_{5\mu m} \cong 2.5\ Q_{2\mu\mathrm{m}}$

(Equivalent to thickness ratio)



Integral of total charge (collected charge)



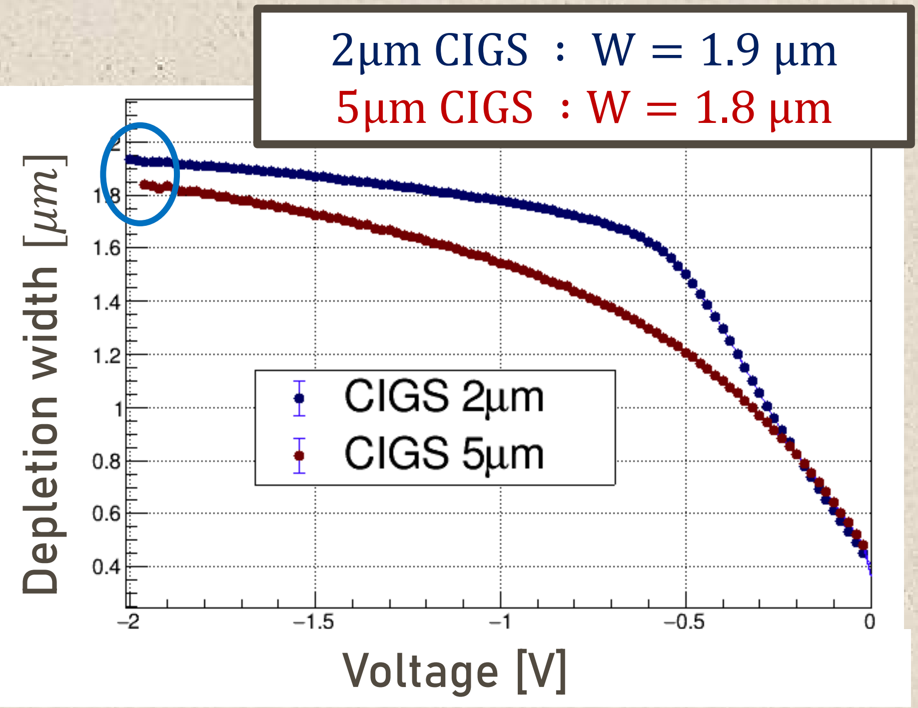


1. Study of Thickness dependence of CIGS (depletion width measuremnt)

Amount of charge collected is proportional to depletion layer thickness $\mathrm{Q_{det}} \propto \mathrm{W}$

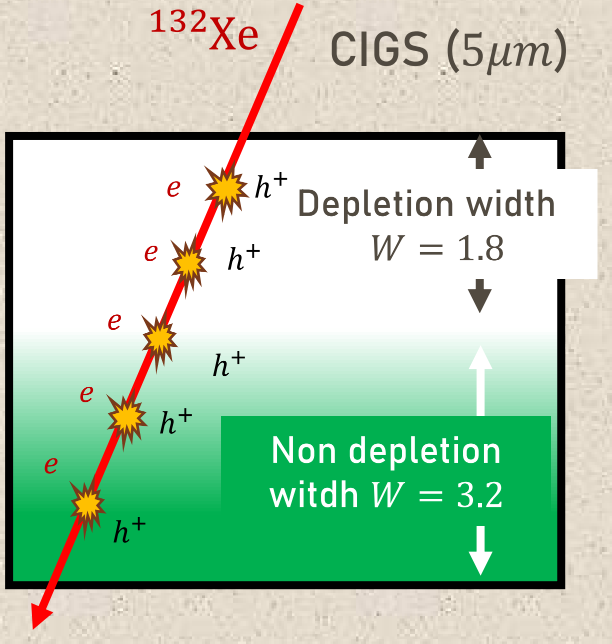
Depletion width (W) can be obtained by capacitance (C_j) measurement

$$C_j \equiv dQ/dV = dQ/(WdQ/\varepsilon_s) = \varepsilon_s/W \quad [W : \text{depletion width}, \varepsilon_s\text{: permittivity } (= 13.5\varepsilon_0)]$$

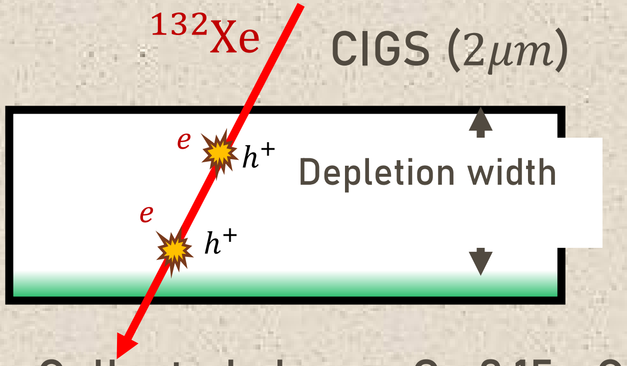


$2\mu\mathrm{m}$ CIGS : $\mathrm{W} = 1.9\ \mu\mathrm{m}$

$5\mu\mathrm{m}$ CIGS : $\mathrm{W} = 1.8\ \mu\mathrm{m}$



Collected charge Q= 0.42 pC



Collected charge Q= 0.15 pC

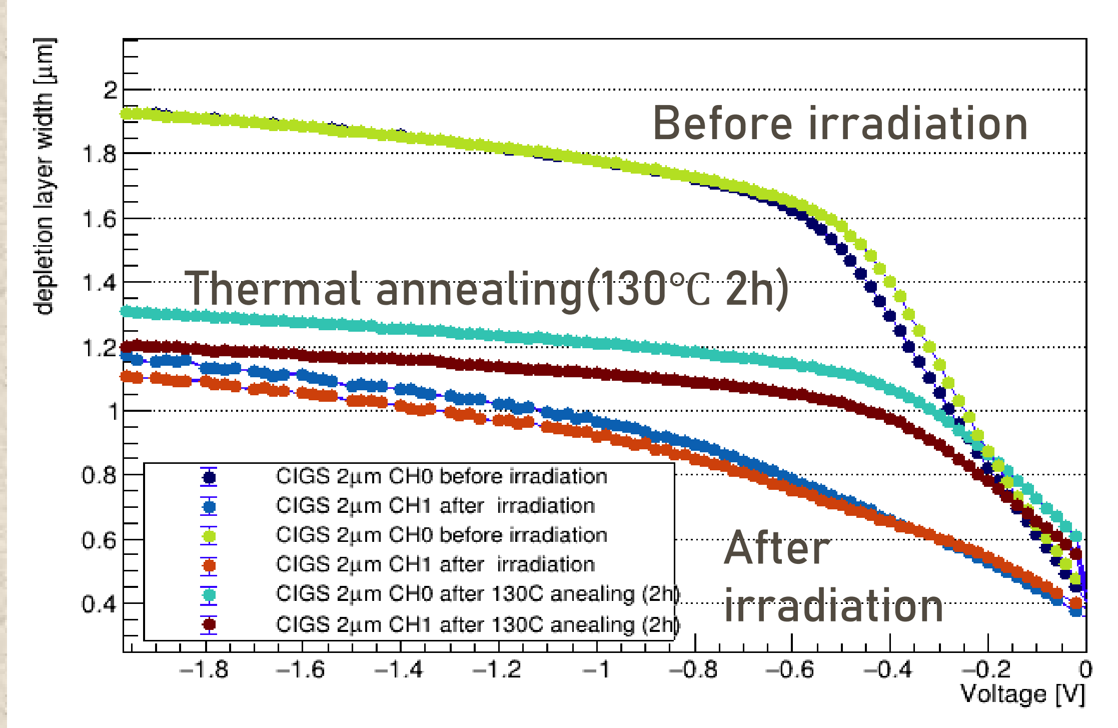
Why can 5 μm CIGS detector collect charge in non-depletion layer ??

Evaluation of depletion layer width after irradiation and thermal annealing

The depletion layer width can be obtained by capacitance measurement

$$C_j \equiv dQ/dV = dQ/(WdQ/\varepsilon_s) = \varepsilon_s/W$$

$$[W\text{ : depletion width, } \varepsilon_s\text{: permittivity } (= 13.5\varepsilon_0)]$$

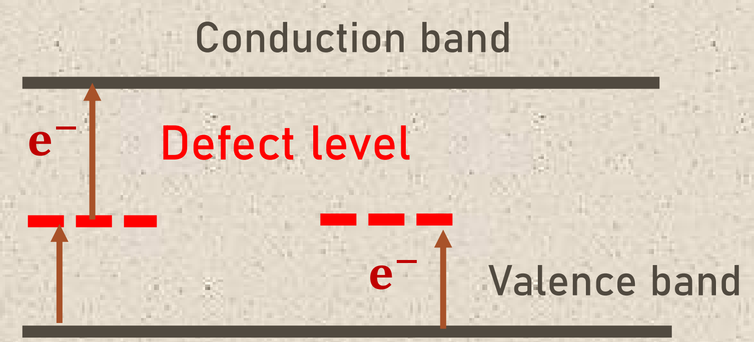


	CH	Before irradiation	After irradiation (0.8 MGy)	After annealing 130℃, 2h
Depletion width [um] at V=-2V	CH0	1.93 (1)	1.17 (0.61)	1.31 (0.68)
	CH1	1.93 (1)	1.11 (0.57)	1.20(0.62)

After irradiation : comparable with decreasing ratio of collected charge ~ 0.6

After annealing : Not sufficient of recovering

Observation of radiation damage recovery in HIMAC experiment



Conduction band

e^-

Defect level

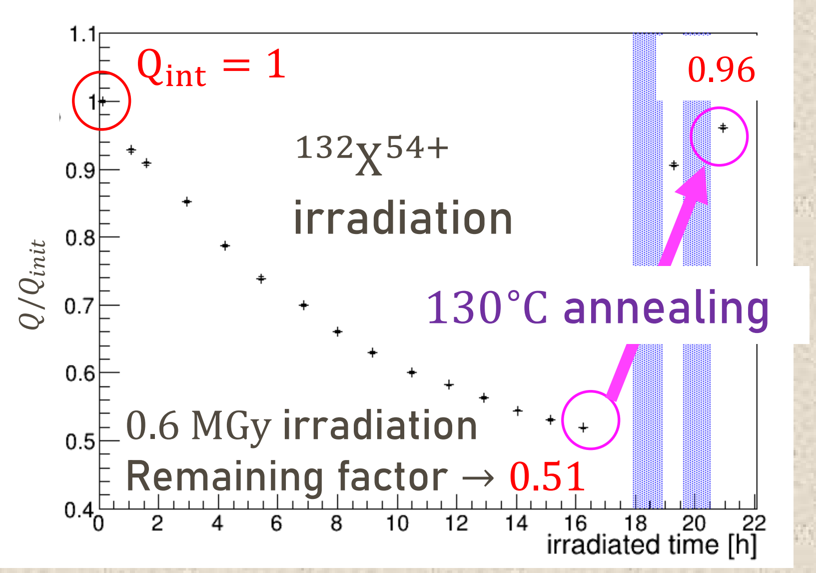
e^-

Valence band

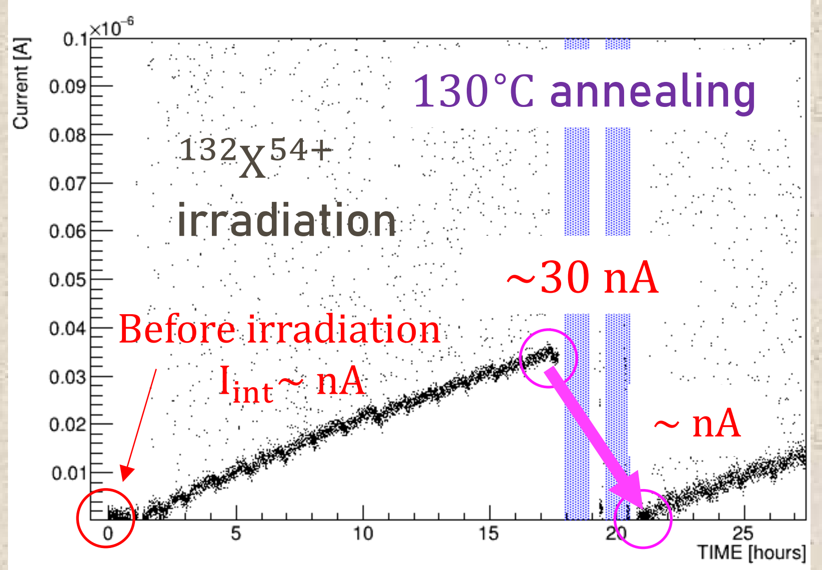
Noise excitation
$I_{rad} > I_{int}$

Charge trap
$Q_{rad} < Q_{int}$

Collected charge monitoring



Leakage current monitoring



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Both collected charge and leakage current were mostly recovered by 130°C annealing (**decreasing defect levels created by radiation damage**).

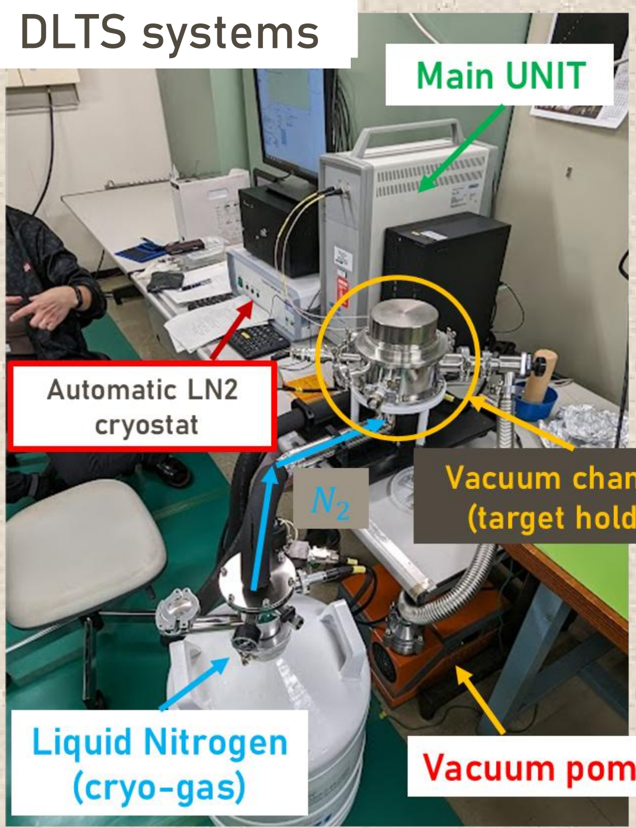
→ **Investigation of defect levels in CYRIC experiment**

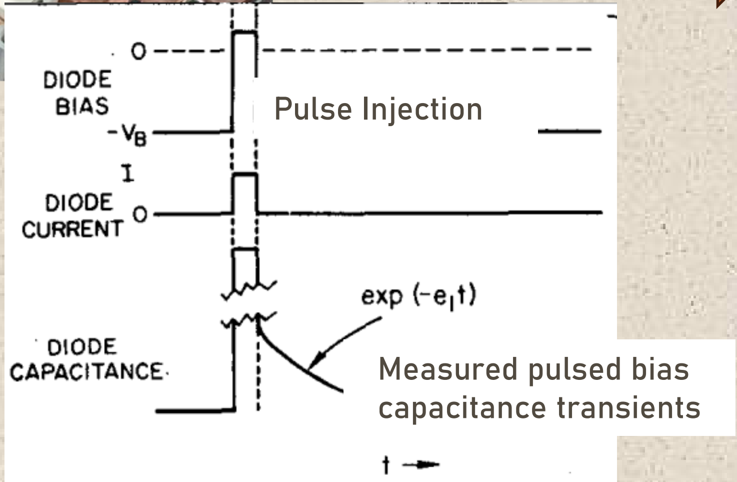
Study of defect levels created proton irradiation (CYRIC)

CYRIC experiment : Irradiated 70 MeV proton to CIGS solar cells $(7.5 \times 10^{15}\ \mathrm{n_{eq}})$

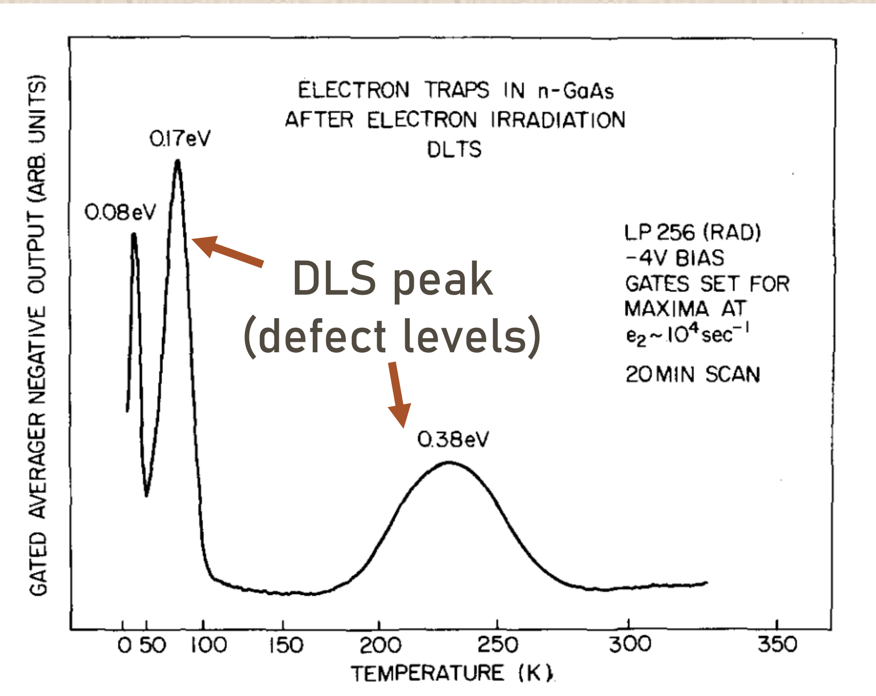
Study of defect levels by Deep Level Transient Spectroscopy (DLTS) method

DLTS systems





Expected results by DLTS method



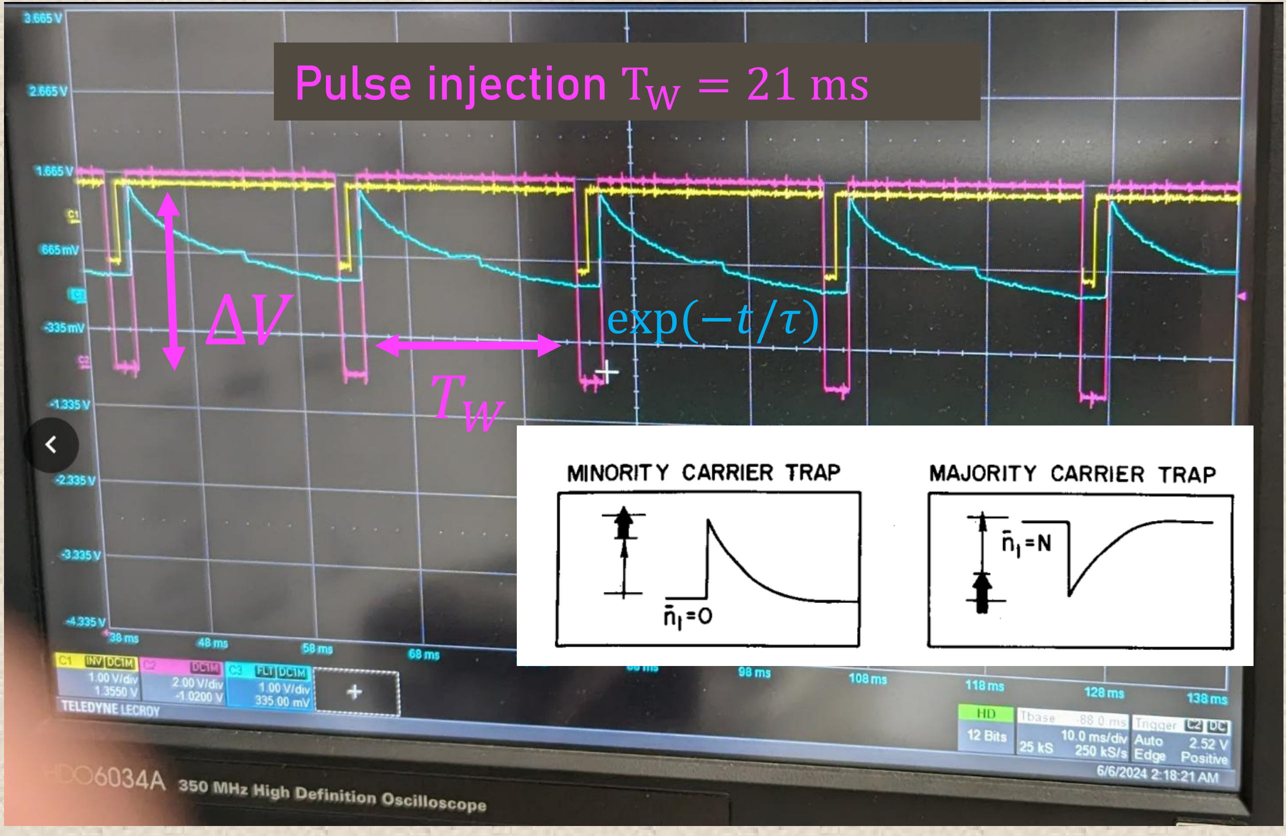
[1] J. Appl. Phys., Vol. 45, No. 7 (1974).

Deep Level Transient Spectroscopy

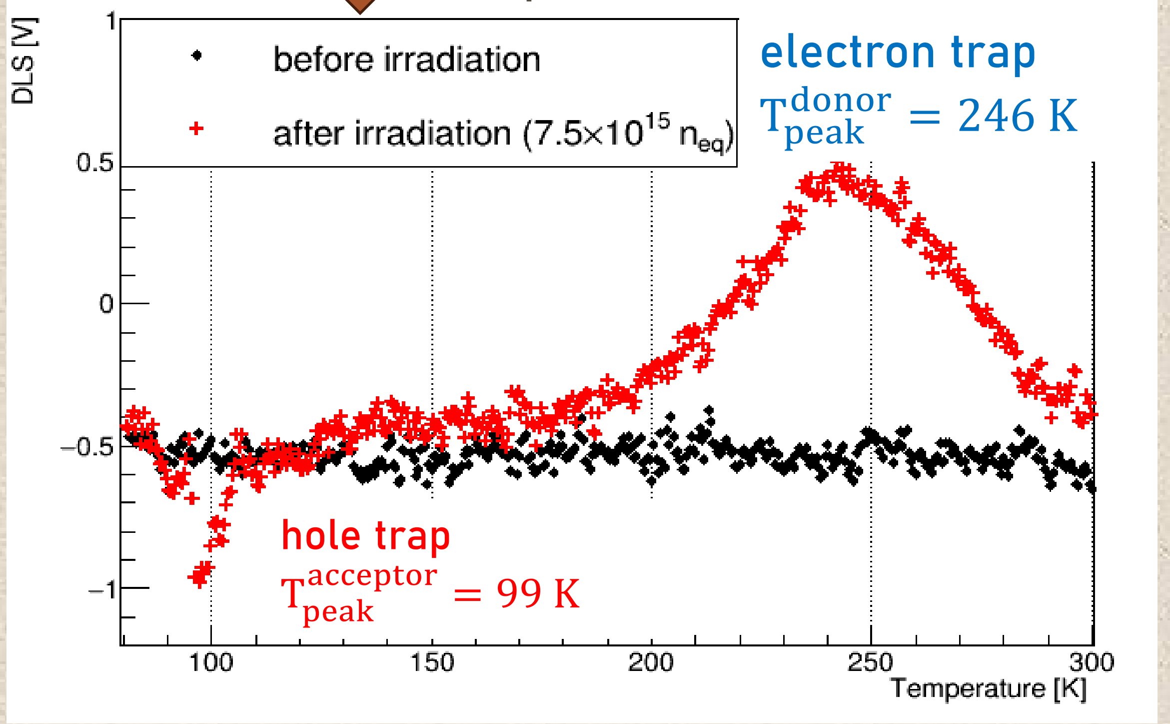
The DLTS signal is created by the lock-in integrator

→ The $\mathrm{T_{peak}}$ was observed by the DLTS thermal scan

$$\mathrm{DLS} = \frac{1}{T_W}\left[\int_0^{\frac{T_W}{2}} e^{-\left(t-\left(t_1+\frac{T_W}{20}\right)\right)/\tau}dt - \int_{\frac{T_W}{2}}^{T_W} e^{-\left(t-\left(t_1+\frac{T_W}{20}\right)\right)/\tau}dt\right]$$



Temperature scan



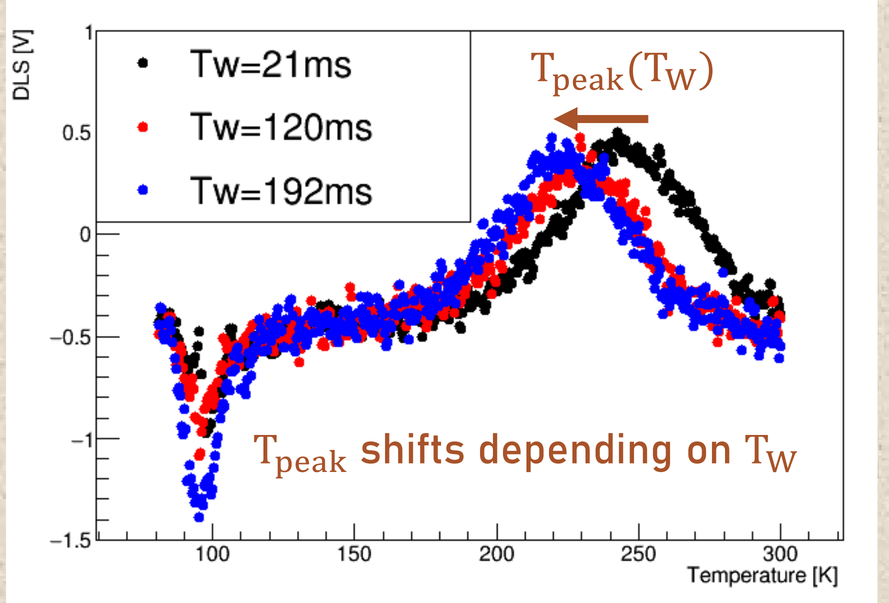
Arrhenius Plot

Trap energy level E_t can be obtained by Arrhenius plot fitting

Arrhenius plot

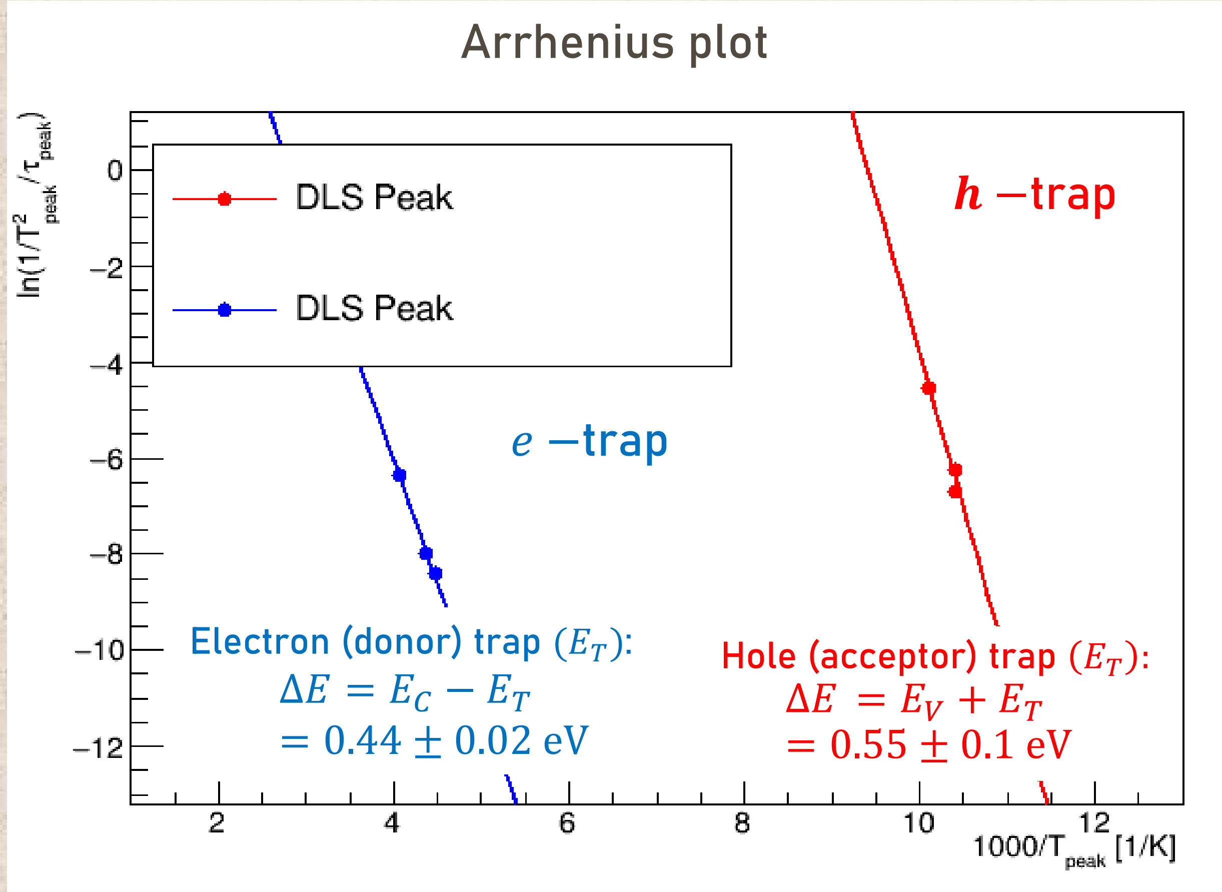
$$\ln\left(\frac{2.17}{T_{\mathrm{peak}}^2 T_W}\right) = \ln(K\sigma_n) - \frac{E_C - E_t}{kT} \quad (K : \mathrm{const.})$$

variable values E_t , σ_n



$T_{peak}(T_W)$

T_{peak} shifts depending on T_W


Arrhenius plot

h −trap

e −trap

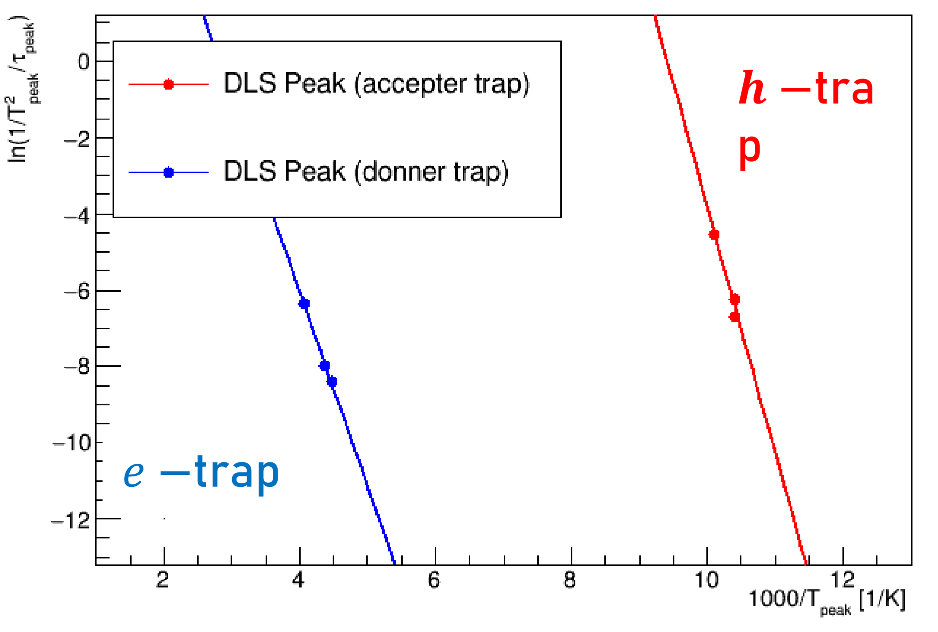
Electron (donor) trap (E_T):
$\Delta E = E_C - E_T$
$= 0.44 \pm 0.02$ eV

Hole (acceptor) trap (E_T):
$\Delta E = E_V + E_T$
$= 0.55 \pm 0.1$ eV

Candidate of defect levels created by proton irradiation

Electron (donor) trap (E_T):
$\Delta E = E_C - E_T = 0.44 \pm 0.02 \text{ eV}$

Hole (acceptor) trap (E_T):
$\Delta E_V + E_T = 0.55 \pm 0.1 \text{ eV}$



Trap defect	Defect level [1]	Electrical properties
V_{Cu}^{-}	$E_V + 0.03$	acceptor
V_{In}^{-}	$E_V + 0.17$	acceptor
V_{In}^{2-}	$E_V + 0.41$	acceptor
V_{In}^{3-}	$E_V + 0.67$	acceptor
In_{Cu}^{2+}	$E_c - 0.34$	donor
In_{Cu}^{1+}	$E_c - 0.25$	donor
Cu_i^{+}	$E_c - 0.20$	donor

Hole trap candidates V_{In} defects

electron trap candidates In_{Cu} defects

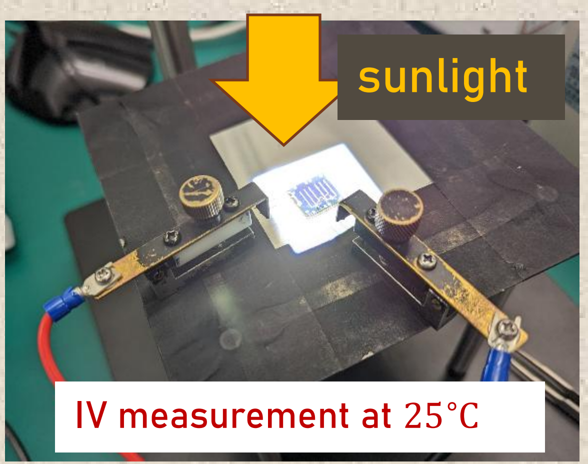
[1] A. Zunger et al., 26th IEEE Phytovoltaic Specialists Specialists Conf. (1997).

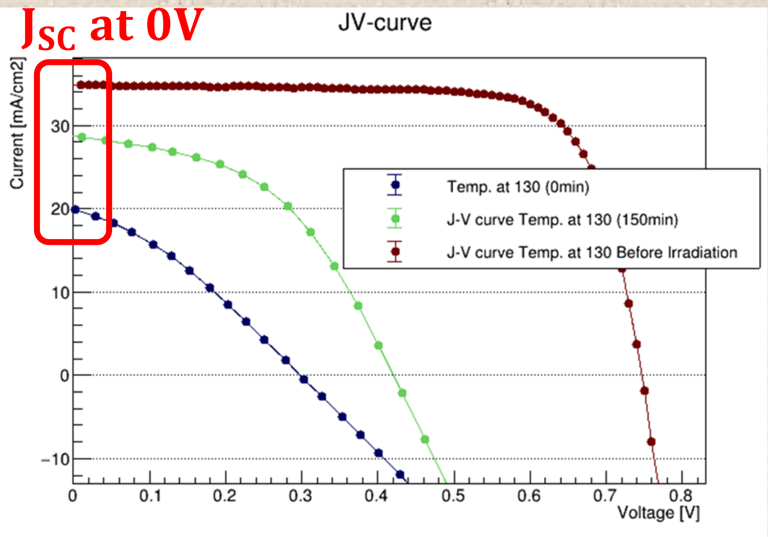
Study of recovery mechanism

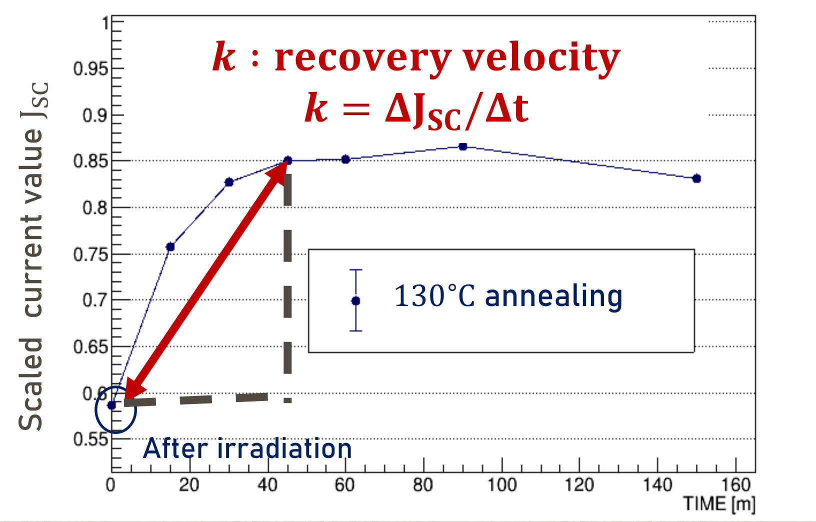
In the chemical reaction ($\mathrm{X} + \mathrm{YZ} \leftrightarrow \mathrm{XY} + \mathrm{Z}$), the correlation between excitation energy E_a and reaction velocity constant k were described by Arrhenius equation as

$$k = A \exp\left(-\frac{E_a}{k_B T}\right) \quad (k_B : \ \text{Boltzmann const.}, \qquad \text{A : const.}).$$

By measuring an excitation energy, the activated atoms during thermal annealing can be identified.







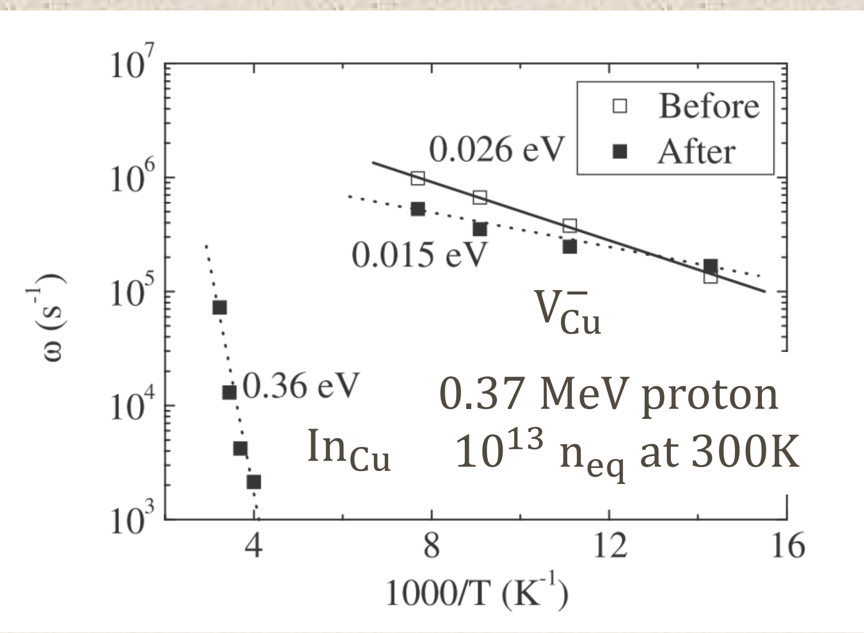
doi : https://doi.org/10.1016/j.nima.2024.169637. https://doi.org/10.1016/j.nima.2024.169637.

Discussion

In the previous study, CIGS solar cells with 0.37 MeV proton irradiation at room temperature (fluence : $\sim 10^{13}\ \mathrm{cm}^{-2}$) had **$V_{Cu}^{-}$ and In_{Cu} defects** [1] .

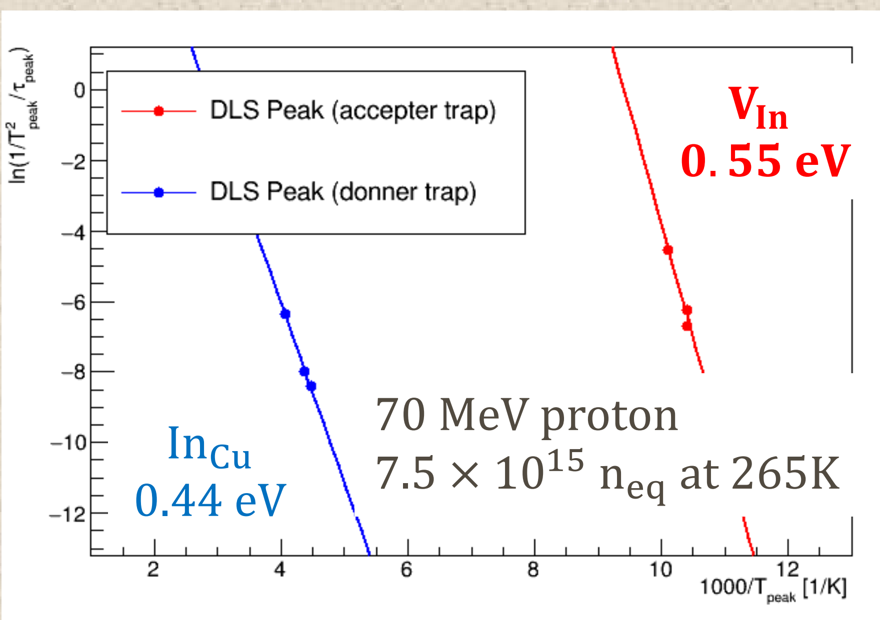
On the other hand, DLTS method observed **In_{Cu} and V_{In} defects** after 70 MeV proton irradiation at -15°C (fluence $\sim 7.5 \times 10^{15}\ n_{eq}$).

Arrhenius plots from admittance spectroscopy



[1]Jpn. J. Appl. Phys. 46 27 (2007).

Arrhenius plots from DLTS (our study)



The defect level of V_{Cu} is low ($E \sim 13$ meV).
It has sensitive to lower temperature region (≤ 77K).
But we successfully observed V_{In} defect in DLTS method.