Facility updates for high-resolution and high-accuracy crystallography

Ichiro Inoue & Kensuke Tono

on behalf of SACLA beamline group

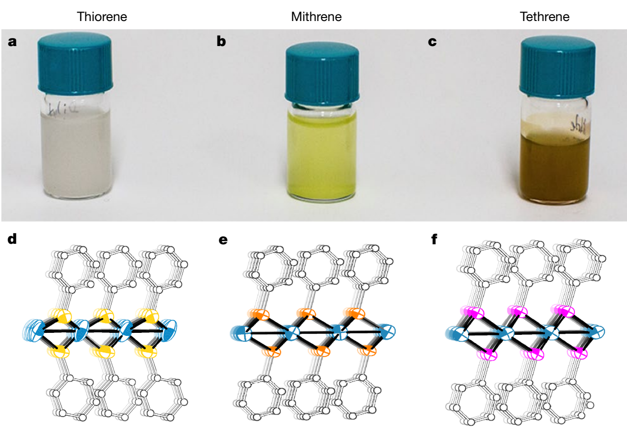
SACLA

RIKEN
Since 1917

Motivation of this breakout session

- Number of users employing high-photon energy XFEL beam (>15 keV) is gradually growing.
- Many interesting outcomes in high-resolution crystallography are being published.

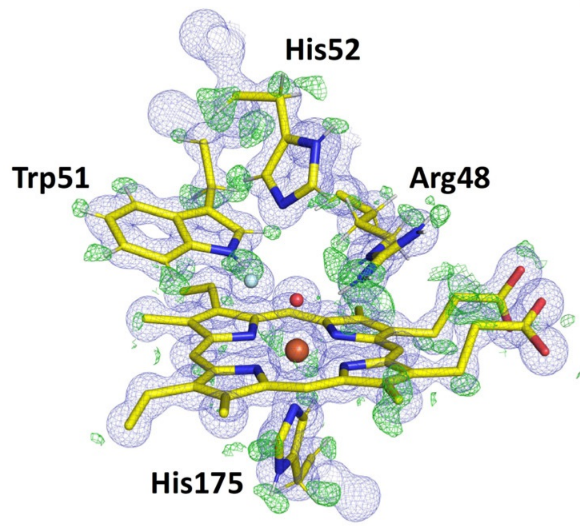
SFX of small molecules



Compound	Thiorene	Mithrene	Tethrene
Formula	$AgSC_6H_5$	$AgSeC_6H_5$	$AgTeC_6H_5$
Molecular weight	217.03	263.93	312.57
Space group	Cc	C2/c	C2/c
a (Å)	7.290	5.938	5.900
b (Å)	5.879	7.325	7.424
c (Å)	28.072	29.202	30.258
α (°)	90	90	90
β (°)	93.80	95.44	97.686
γ (°)	90	90	90

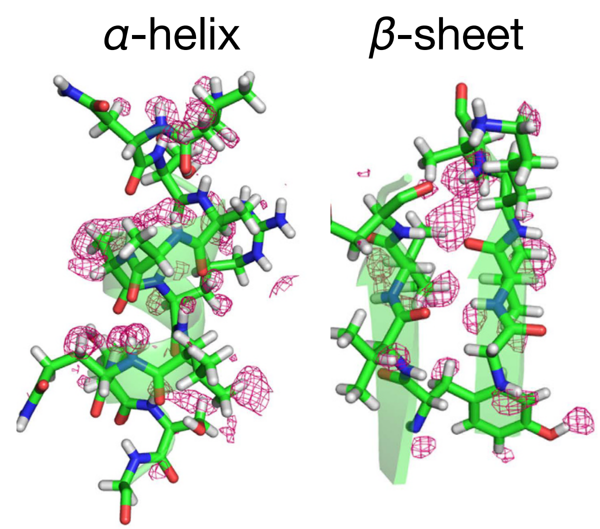
Schriber, Paley, Hohman, *Nature* 601, 360 (2022)

Hydrogen atoms in enzymes



cytochrome c peroxidase

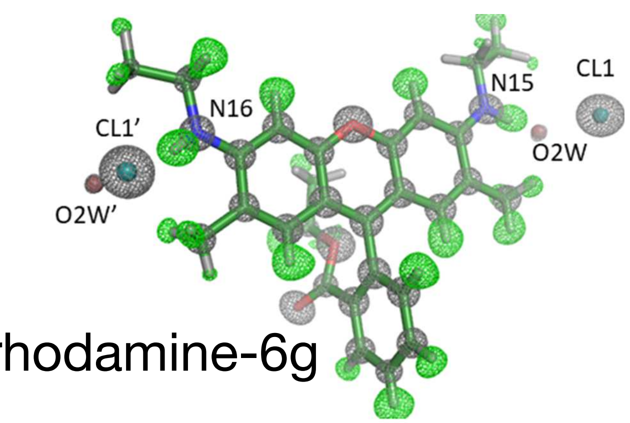
Kwon, Sugimoto, Raven, Moody, *Angew. Chem. Int. En.* 60, 14578 (2021)



proteinase K

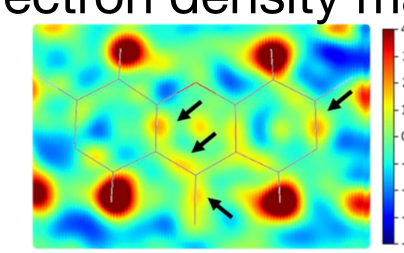
Masuda, Sugahara, *Sci. Rep.* 7, 45604 (2017)

Covalent electrons and hydrogen atoms in a small molecule



rhodamine-6g

Hydrogen omit electron density map



Takaba, Maki-Yonekura, Yonekura, *ChemRXiv* 10.26434/chemrxiv-2021-jvbfl

-> It's a good timing to learn about the latest researches in this field and discuss about future facility capabilities

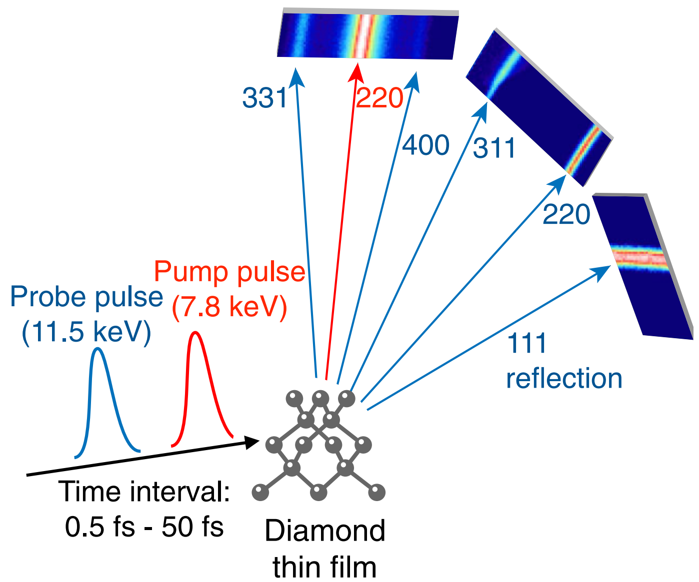
Photon parameters compared to other XFEL facilities

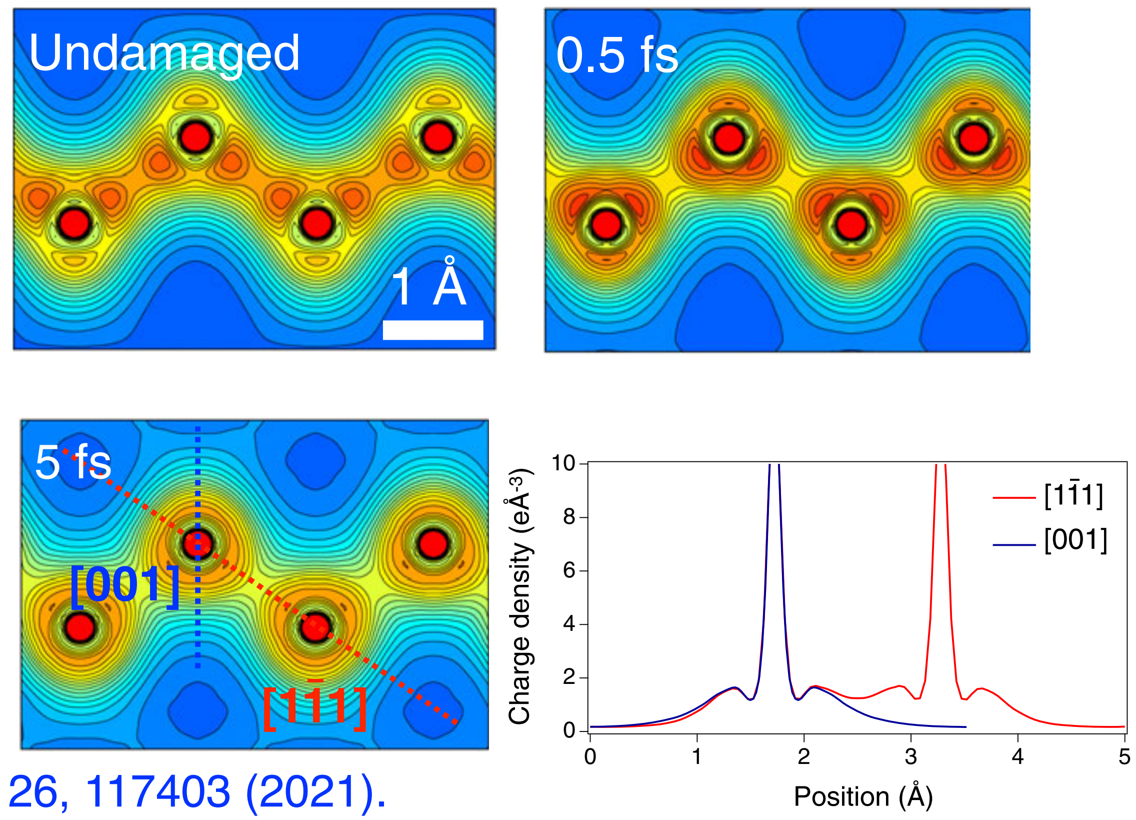
	Photon energy (fundamental)	Pulse energy	Pulse duration (FWHM)	Rep. rate	Unique modes/ capabilities
SACLA	4-22 keV	up to 1 mJ	~6 fs (fixed)	BL2+BL3: 60 Hz in total	•Two-color •Self-seeding •Hight beam stability •Extreme focusing
LCLS	0.25-25 keV	~4 mJ for 30 fs pulse (proportional to pulse duration)	Variable (nominally, 30 fs)	120 Hz (1 MHz @LCLS-II HE)	•Two-color •Self-seeding •Attosecond
European XFEL	5-24 keV	0.5-4 mJ	20-60 fs	2.7 kHz (4.5 MHz for intra-train)	Under commissioning (two-color, self-seed)
PAUL SCHERRER INSTITUT PSI	2-12 keV	~0.5 mJ for 120 fs pulse (proportional to pulse duration)	Variable (nominally, 120 fs)	100 Hz	•Two-color •Large bandwidth
PAL POHANG ACCELERATOR LABORATORY	2-15 keV	~1 mJ	•Variable (10-40 fs)	60 Hz	•Two-color •Self-seeding •Excellent timing stability

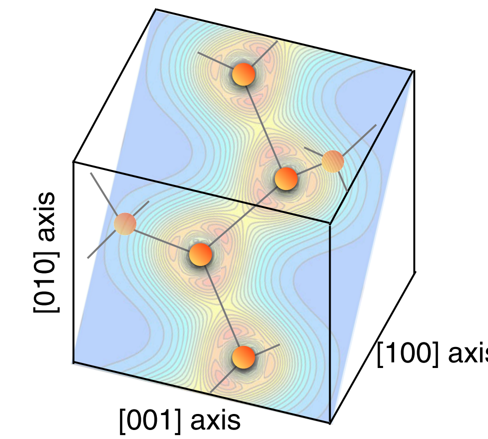
SACLA is highly competitive in high-resolution and high-accuracy crystallography

Sub-10 fs pulses are definitely needed for accurate structure determination

Visualizing charge-density in x-ray excited diamond



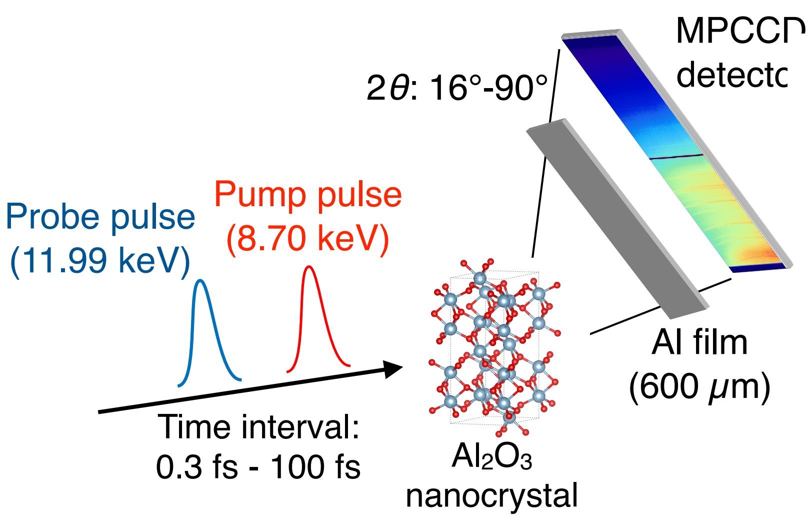


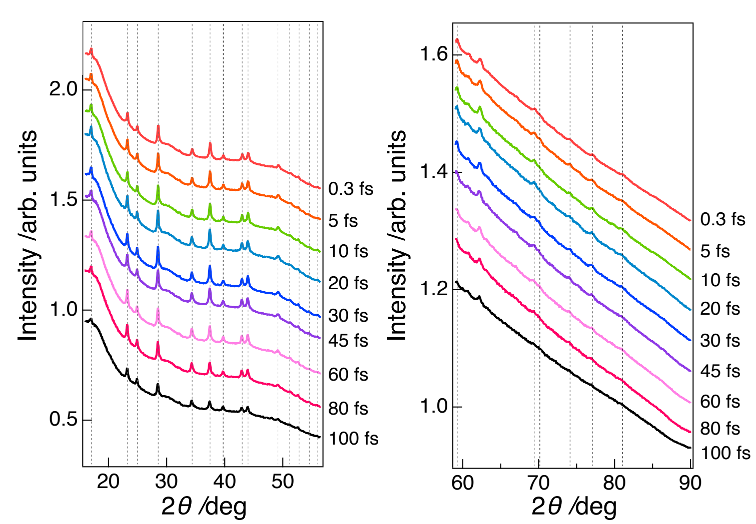


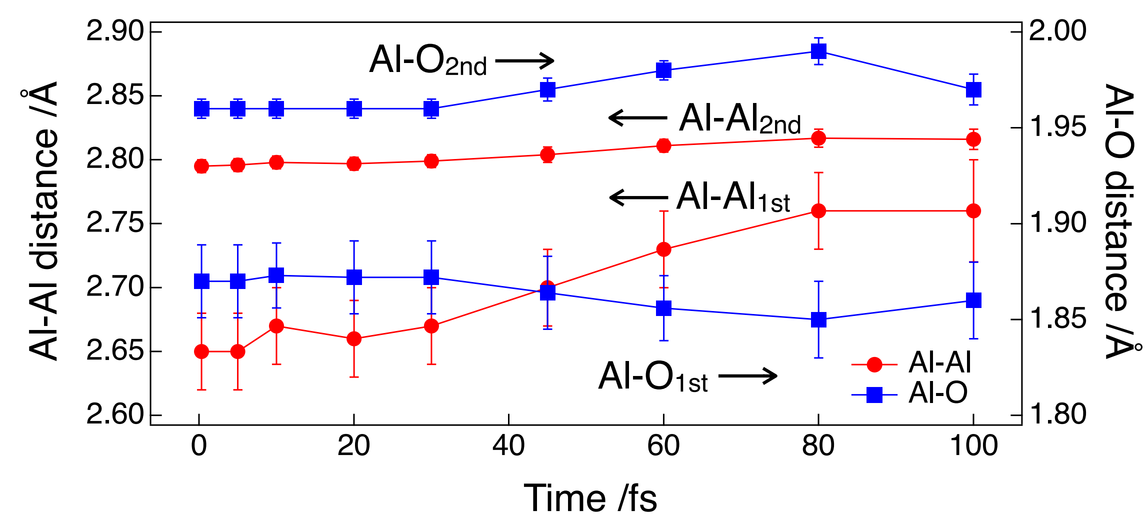
Inoue, *PNAS* 113, 1492 (2016).
Inoue, Kasai, Nishibori, *Phys. Rev. Lett.* 126, 117403 (2021).

Chemical bonds are broken within **5 fs** after irradiation with an X-ray pulse

X-ray pump X-ray probe of Al_2O_3







Inoue, Nishibori, Beata, *arXiv:2112.05430* (2021).

Interatomic distances start to change at **~20 fs** after irradiation with the x-ray pulse

Program

Introduction
15:00-15:20
Ichiro Inoue (SACLA)
‘Facility updates for high-resolution and high-accuracy femtosecond crystallography’

Research highlights and requests to facility
15:20-16:20
Kiyofumi Takaba (RIKEN)
‘Sub-Å resolution SACLA-SFX for organic chemicals’

Hidetaka Kasai (U. Tsukuba)
‘Serial femtosecond crystallography for materials science and chemistry’

Hiroshi Sugimoto (RIKEN)
‘Hydrogen atoms in protein structure: high resolution analysis of heme enzymes’

Discussion (chair: Kensuke Tono)
16:20-17:00

Facility updates

- ☑ New standard configuration setup for high-resolution crystallography (2022B~)
- ☑ Detector updates (2023?)
- ☑ Optics updates at BL2 (2022B~)

New standard configuration setup for high-resolution crystallography

Current standard configurations

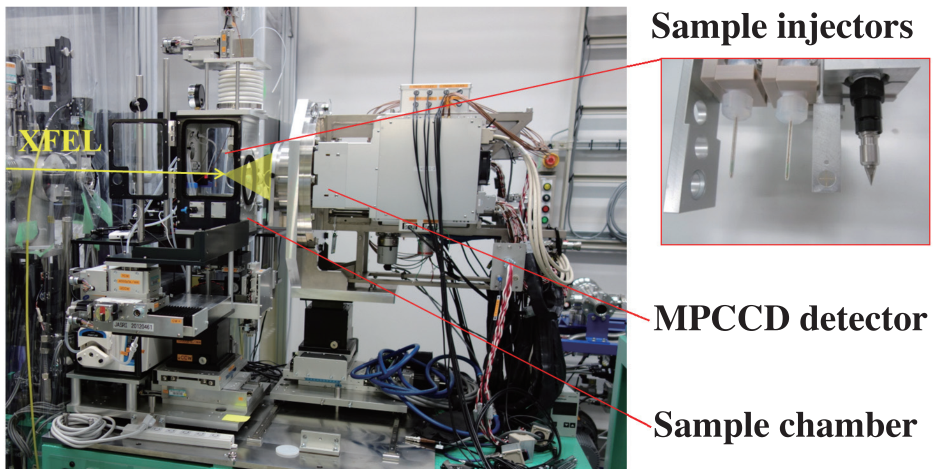
w/ MPCCD (max 2θ: 48 deg)

Camera length: **50 mm**

Direct detection of X-ray photons

(Si 300 μm, can detect single photon)

Small sensor area: **110 mm X 110 mm**



Tono, *JSR* 22, 532 (2015)

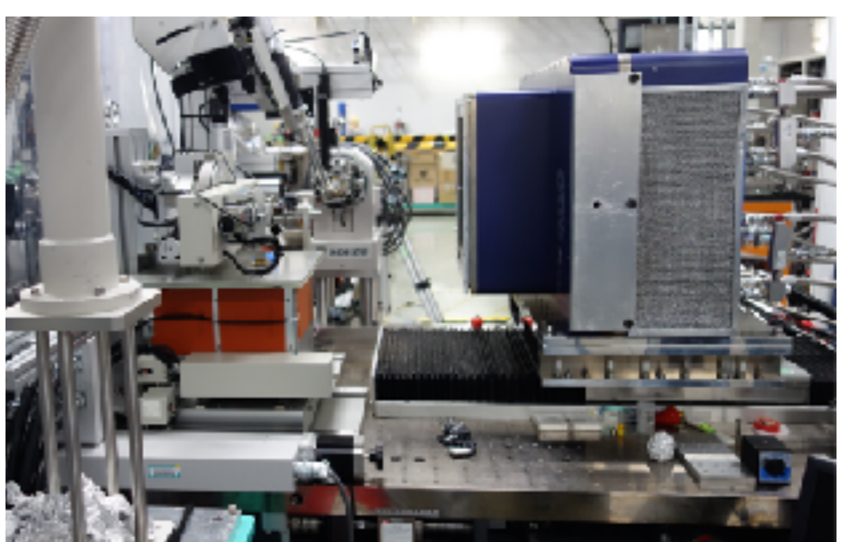
w/ Rayonix 300-HS (max 2θ: 56 deg)

Camera length: **100 mm** or more

Large sensor area: **300 mm X 300 mm**

Phosphor (Gd_2O_2S, 40 um thick) coupled CCD:

good quantum efficiency at high photon energy (*cf.* 90%@15 keV)

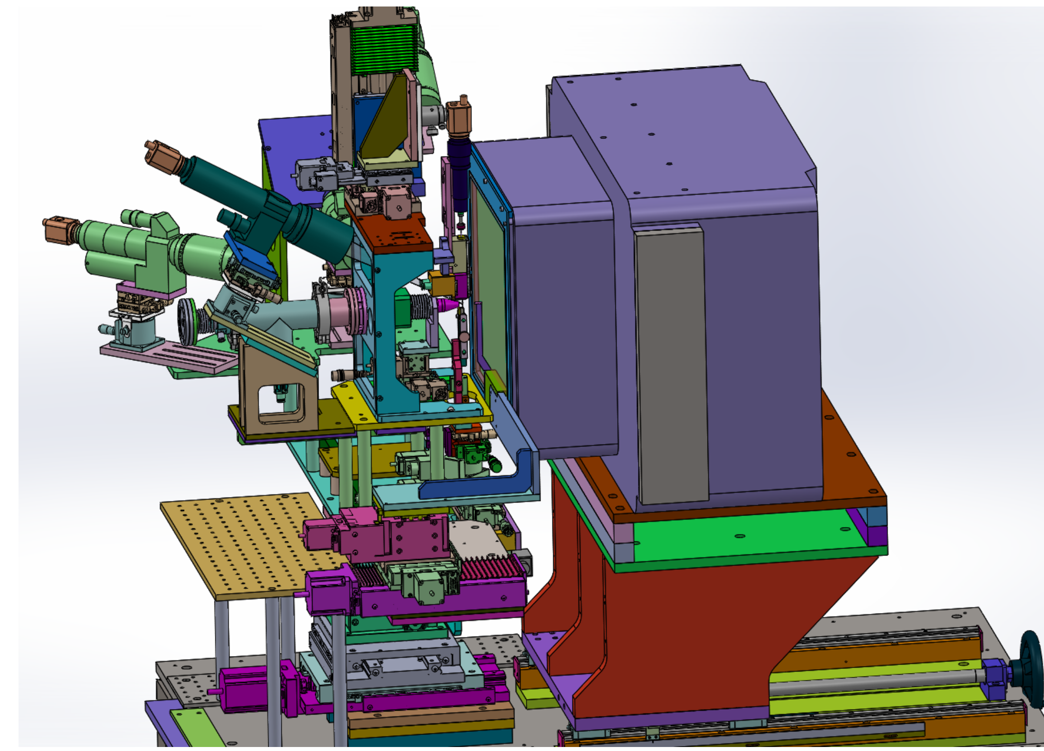


Additional standard configurations (2022B~)

w/ Rayonix 300-HS (max 2θ: 72 deg)

camera length **50 mm** or more

(changeable during beam time)



Resolution limit determined by the detector geometry:

- 0.88 Å @12 keV
- 0.70 Å @15 keV
- 0.54 Å @20 keV

cf. IUCr cif check:

s>0.59 $Å^{-1}$, *i.e.*, d_{min} <0.85 Å

Detector development

Si-based direct illumination detector (CITIUS) will be introduced as a successor of MPCCD (probably next year for the 20.2 M variant, but the schedule has not been fixed yet due to the recent semiconductor crisis)

※ MPCCD is available for a while, but will be replaced to CITIUS in a long term.

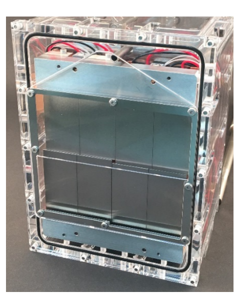
Parameters		Value: CITIUS XFEL for SACLA	Ratio	Value: MPCCD Phase III/III-L	Unit
Sensor	Thickness	650	← x2.1	300	μm
	Pixel Size	72.6 μm	← x1.5	50	μm
	Pixel Number	0.28	← x0.6	0.5	Mpixel/sensor
	Image Format	384 x 728		512 x 1024	pixels
	Noise	60 e- or better		300/60	e-rms
	Peak Signal	17,000	← x7	2,400	Phs.@ 6 keV
	Frame Rate	60 (max. 5 kHz)		60 or 30 Hz	
Largest System	Pixel Number	20.2 Mpixel	← x5	4	Mpixel
	Image Area	325 x 363		100 x 100	mm^2
Tiling	Inter-module gap	~3 mm	← x3	< 1 mm	

CITIUS: Camera Head Variants (under development)

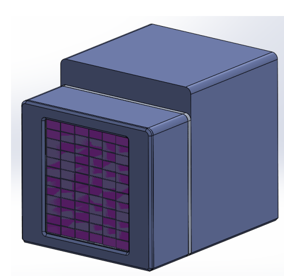
280k Compact

36 x 96 x 197 mm^3
~1 kg
Fan-less operation is possible.

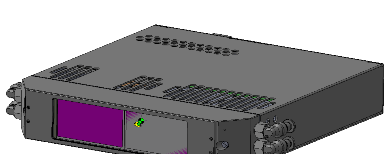
2.2M standard



20.2M standard



560k (Linear) Compact



SACLA goes to higher repetition operation in long term, MPCCD need to be replaced by CITIUS.
However,
- not all the spec. will be better.
- replacement demands significant budget and internal resources.

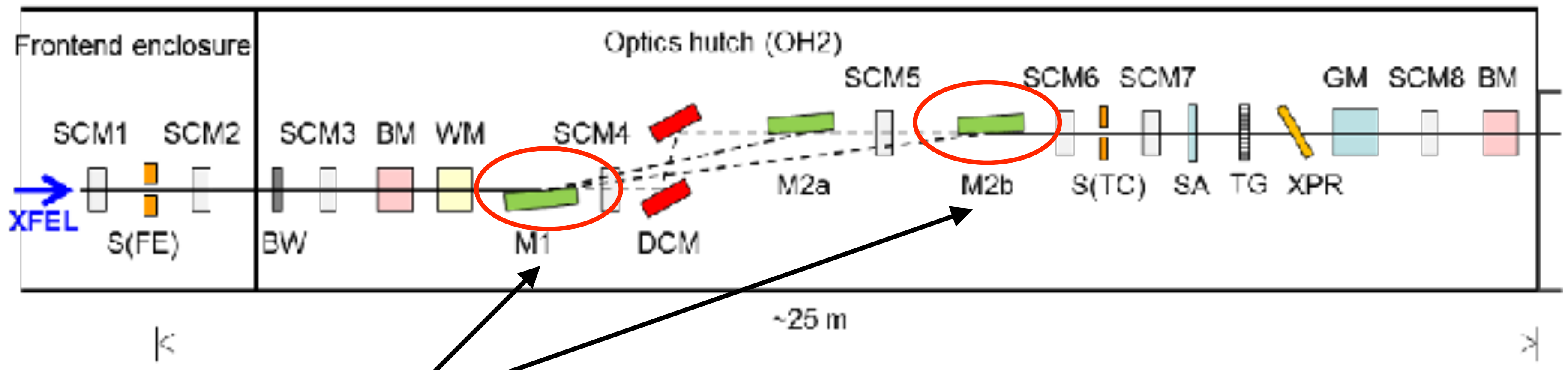
We would like to have user comments, especially, on
- Deployment Priority to CITIUS detector
- MPCCD detector not replaceable by CITIUS

We are finalizing deployment plan for FY2022-2023 by the end of April 2021.

- Large sensor area (325 mm X 363 mm)
- Single photon sensitivity
- Large dynamic range
- Inter-module gap as large as 3 mm
- Pixel size is a bit larger than that of MPCCD

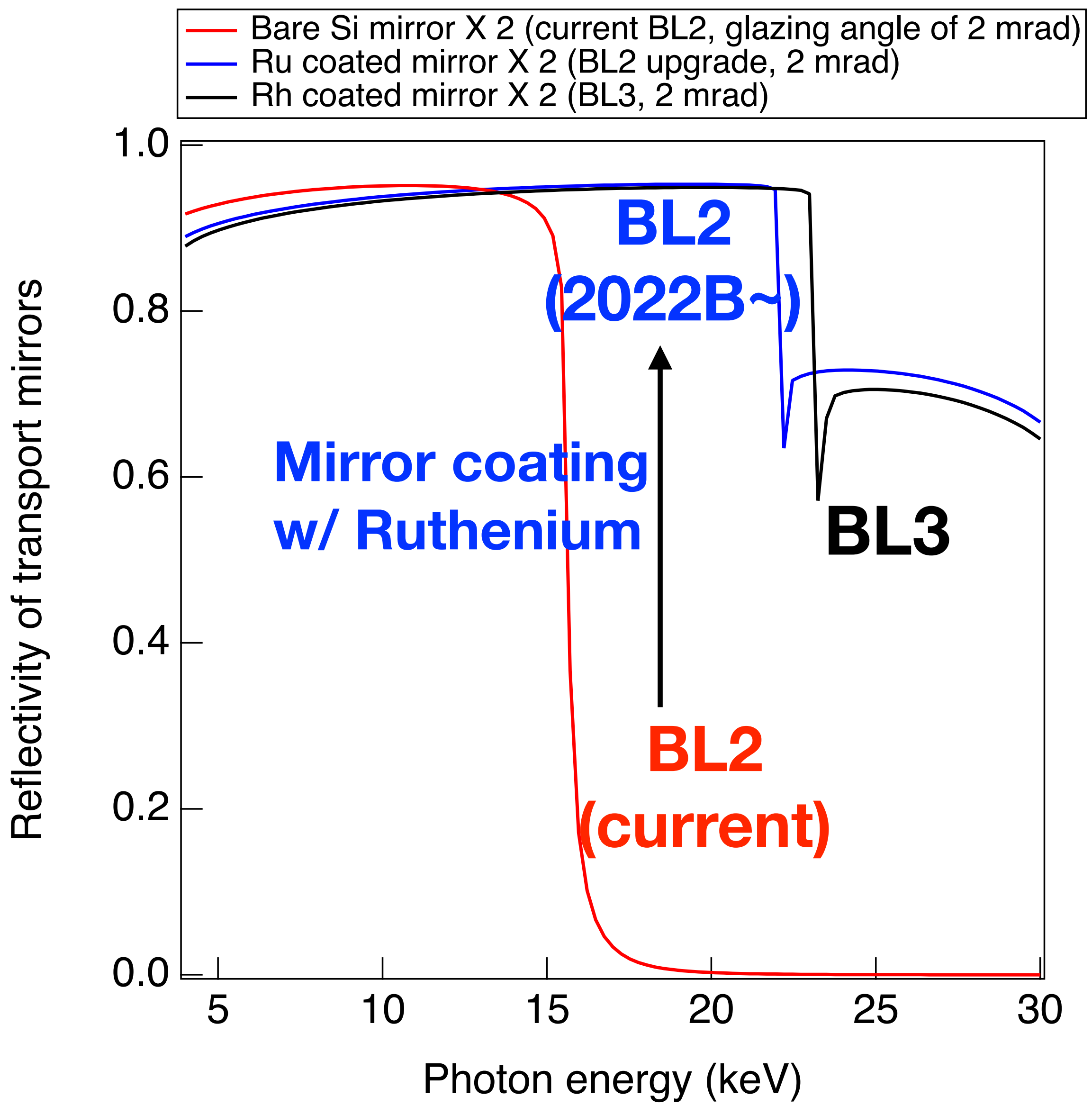
Slides from Takaki Hatsui (RIKEN)
SACLA Users' meeting 2021

Optics development for high photon energy operation (BL2)



Expand available photon energy range of BL2 by coating mirrors

High photon energy (up to 22 keV) will be available at BL2 from 2022B



cf. Specification of current mirror parameters

Plane mirror for beam transport (a)

	Original	Current
Mirror length	0.4 m	0.5 m (BL3)/0.6 m (BL2)
Glancing angle	4 mrad	4 mrad
Acceptance	1.6 mm	2.0 mm (BL3)/2.4 mm (BL2)
Coating	None (Si)	Rh/Si
Cutoff energy	7.5 keV	16 keV (Rh) /7.5 keV (Si)

Plane mirror for beam transport (b)

	Original	Current
Mirror length	0.4 m	0.5 m (BL3)/0.6 m (BL2)
Glancing angle	2 mrad	2 mrad
Acceptance	0.8 mm	1.0 mm (BL3)/1.2 mm (BL2)
Coating	None (Si)	Rh/Si
Cutoff energy	15 keV	33 keV (Rh) /15 keV (Si)

Tono, *Proc. SPIE* 10237 (2017).

Summary and discussion points from facility point of view

Facility update plans

- ☑ New standard configuration setup for high-resolution crystallography (2022B~)
- ☑ Detector updates (2023?)
- ☑ Optics updates at BL2 (2022B~)

-We would like to have feedback from users and any kinds of suggestions for moving high-resolution fs crystallography forward

-> Discussion part in this session