Nanoscale subsurface dynamics of solids upon high-intensity laser irradiation observed by femtosecond grazing-incidence x-ray scattering

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and
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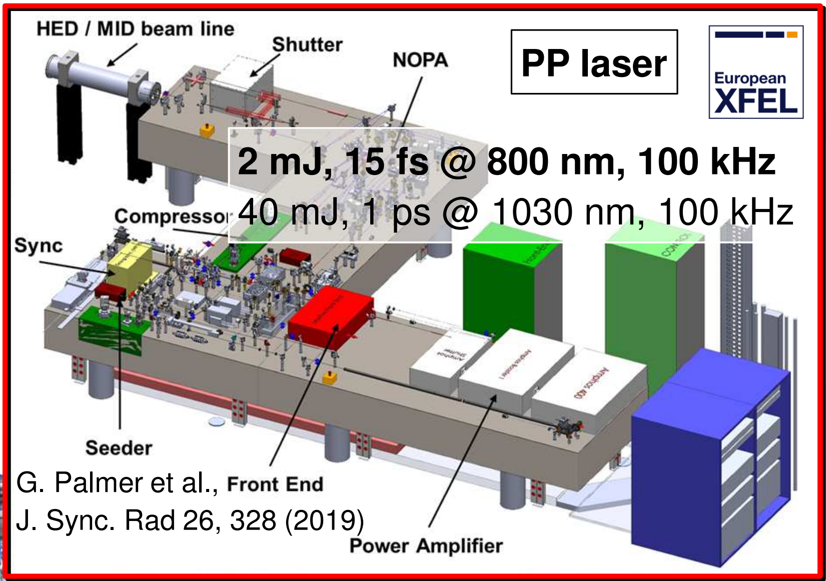
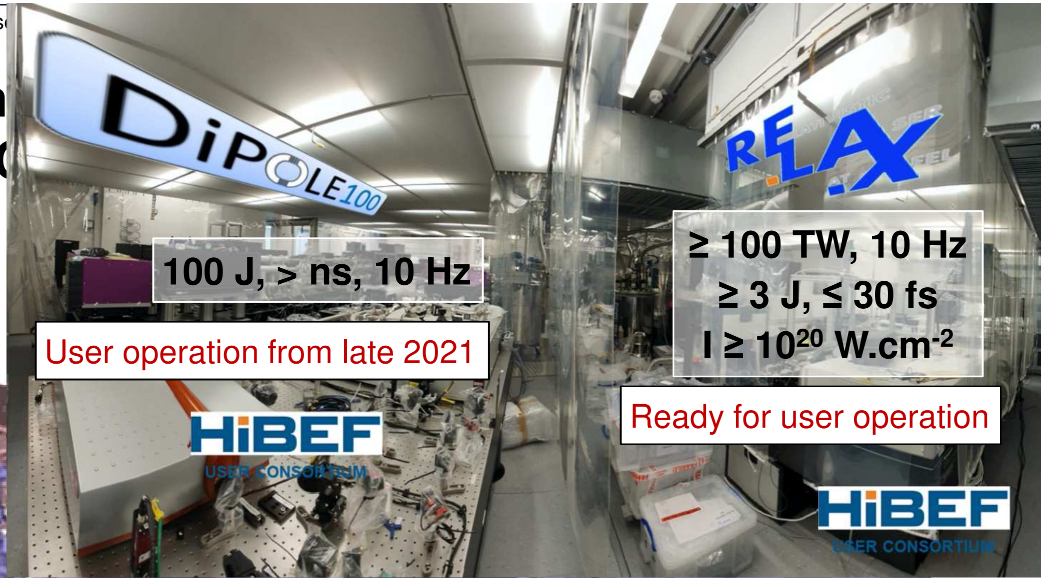
27th Jan. 2021
European XFEL, Users' meeting

Outline

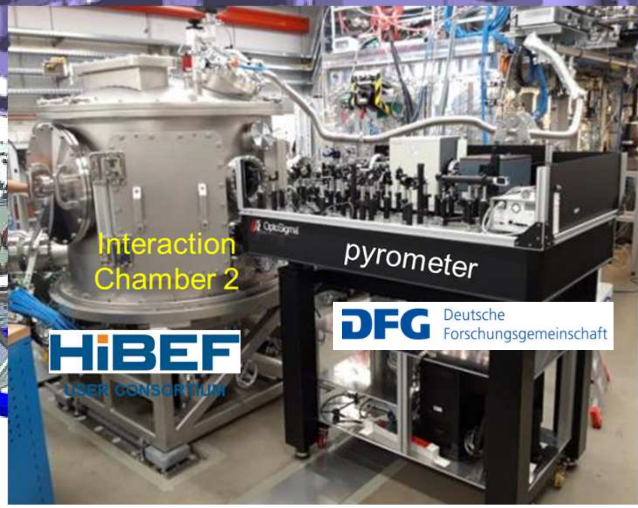
- Introduction
- Experiments at SACLA XFEL (2018 Nov. and 2020 Feb)
  - "High-resolution characterization of high-intensity laser interaction with dense-plasmas using time-resolved grazing-incidence small-angle x-ray scattering"
- Experiment at EuXFEL, HED instrument (2020 Oct.)
  - Proposal #2716: "Probing of phase transition kinetics at the surface of femtosecond laser-heated warm-dense gold with grazing-incidence x-ray diffraction"
  - First user experiment using the PP laser at HED
    - ► Pulse arrival monitor (PAM)

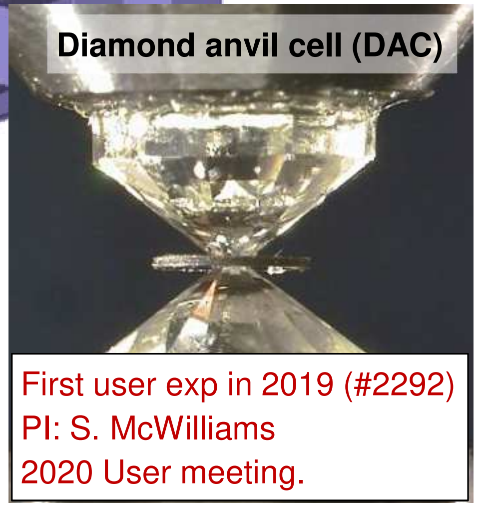
men
& IC





IC1

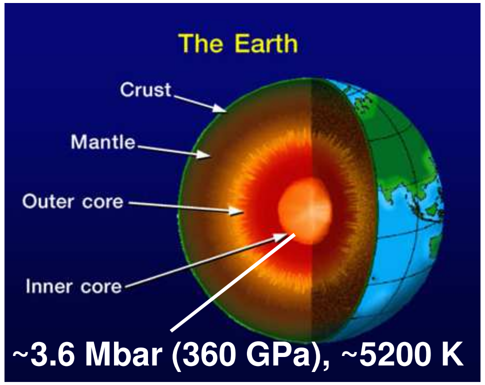


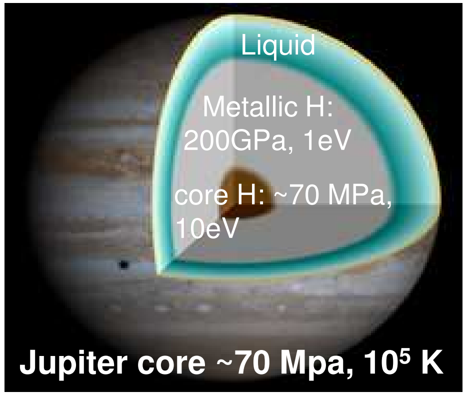


X-ray

European XFEL

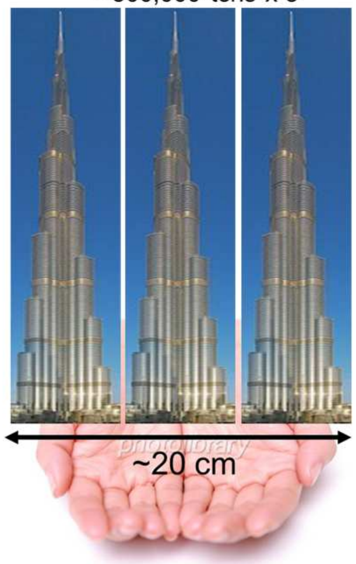
Creating and probing a high-energy-density states – planetary science, bright and compact particle/radiation sources, and more



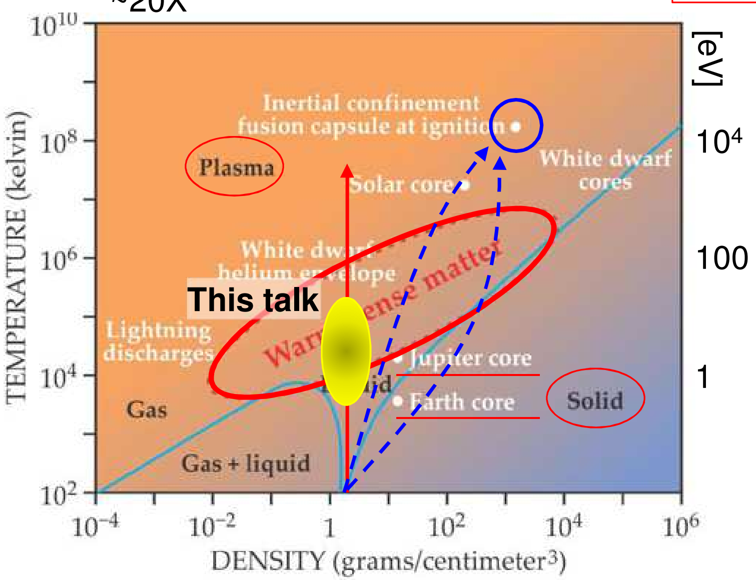


~20X

Burj Khalifa (Dubai)
~ 500,000 tons x 3



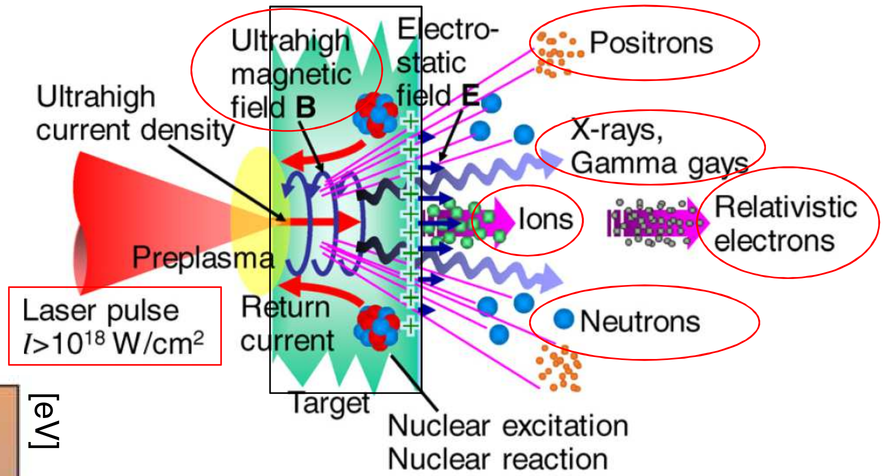
Same



ean XFEL

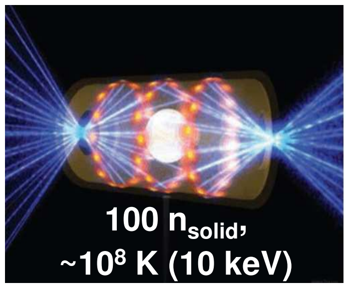
https://physicstoday.scitation.org/do/10.1063/pt.5.7158/full/

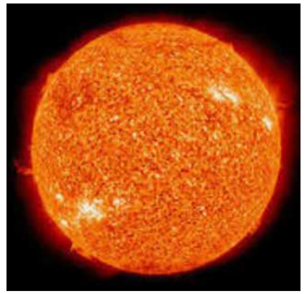
Very high-intensity laser interaction with solids
➔ compact, bright particle / radiation sources



Daido, Nishiuchi and Pirozhkov Rep. Prob. Phys. 75, 056401 (2012)

Fusion energy (ICF)

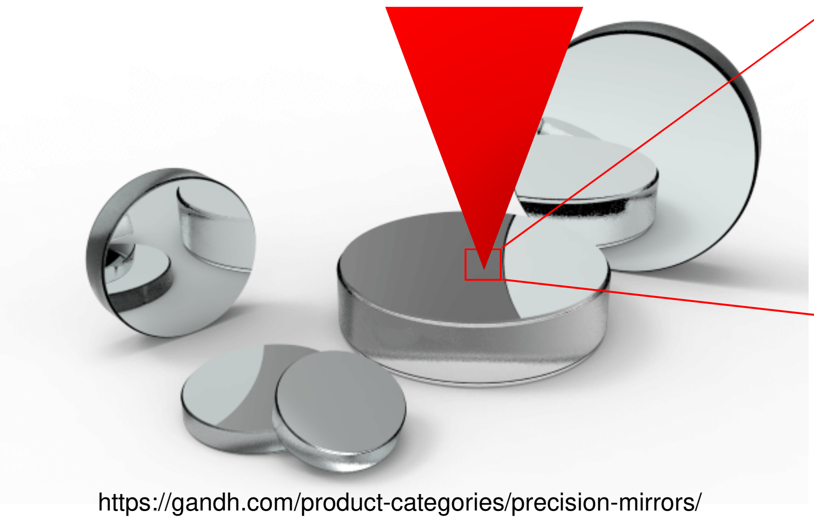




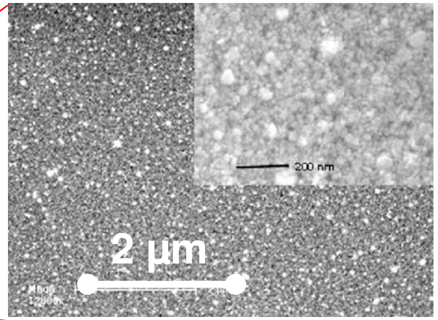
Science, 327, 514 (2010)

"Surface"

Femtosecond laser (e.g., ≥ 10^{12} W.cm^{-2})

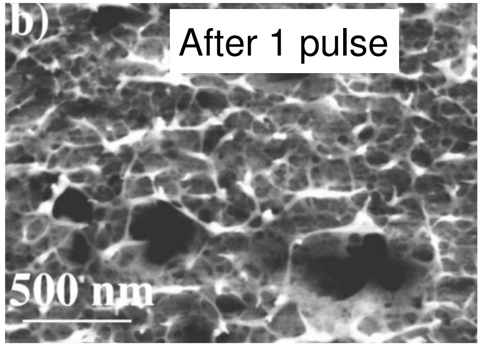


https://gandh.com/product-categories/precision-mirrors/



M. Zaier et. al., Sci. Rep. 7: 12410 (2017)

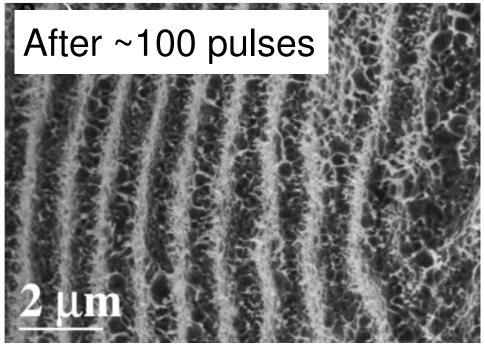
Femtosecond laser ablation



Nano-structure appears

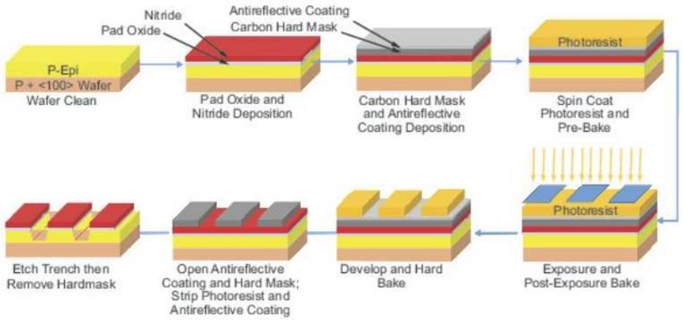
LIPSS (laser-induced periodic surface structure)

Nano-gratings

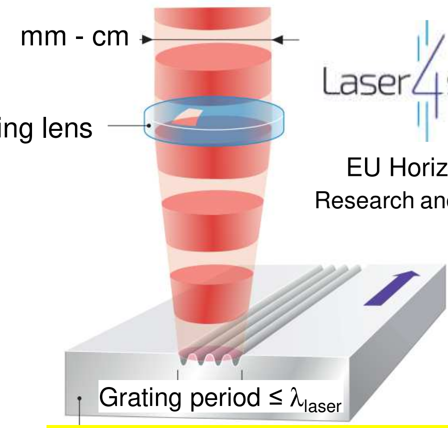


A. Y. Vorobyev and C. Guo PRB 72, 195422 (2005).

Lithography → several step process



From: https://www.newport.com/n/photolithography-overview



Laser4surf

EU Horizon 2020
Research and Innovation

J. Bonse and S. Gräf, Las. Phot. Rev. **14**, 2000215 (2020).
A.Y. Vorobyev and C. Guo, Laser Photon. Rev., **7**, 1, (2012)
...

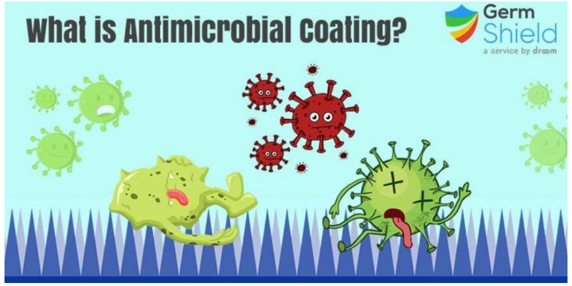
Femtosecond laser is a tool for nanostructuring of materials

https://www.laser4surf.eu/wp-content/uploads/2019/01/Laser-comparing.pdf

Nanoscale surface morphology plays decisive roles in versatile fields

Damaging bacteria

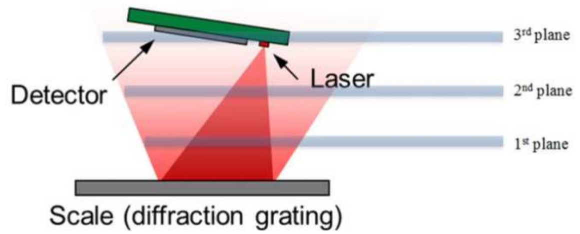
by producing reactive oxygen species



From: https://blog.droom.in/antimicrobial-coating

Optical encoder:

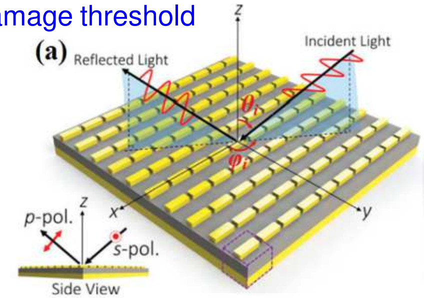
Nanostructured tape optimizes the reflectivity for more accurate measurement



From: https://www.celeramotion.com/microe/optical-encoders/

Advanced optics

Subwavelength surface structure acts as optical anisotropic media ➔ reflective waveplate with higher damage threshold



Shugi Chen et al., DOI: 10.5772/66036

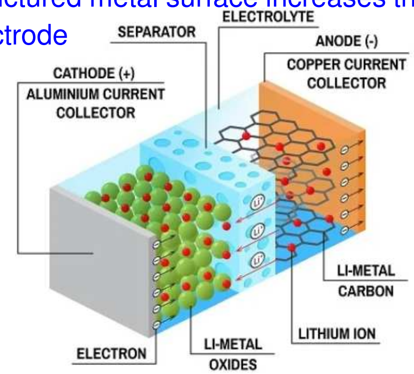
Dental implant, screws:

Structured surface bonds much better with natural bones

From: https://www.bickfordandshirley.com/blog/expect-process-dental-implants-in-hiram/

Battery

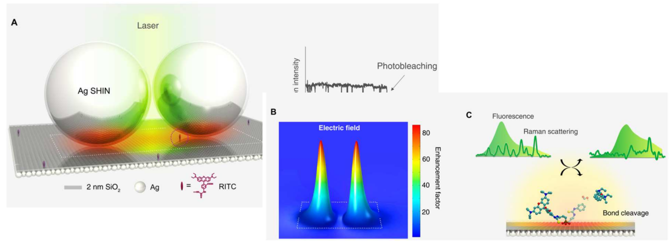
Structured metal surface increases the capacity of the electrode



From: sivVector/Shutterstock.com

Local field enhancement

Plasmon enhanced spectroscopy

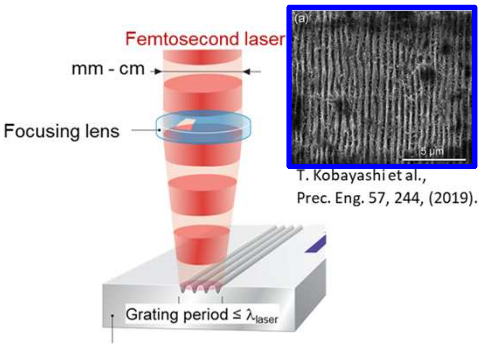


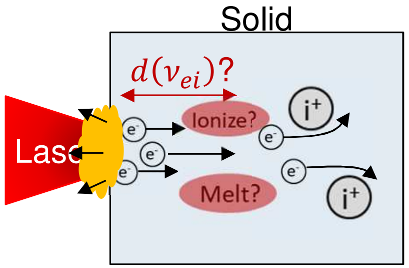
Sci. Adv. 6, eaba6012 (2020)

Nanostructured surface can absorb lasers efficiently ➔ create higher HED state with a given laser system

Most of these application examples are from: Laser4surf EU Horizon 2020 Research and Innovation

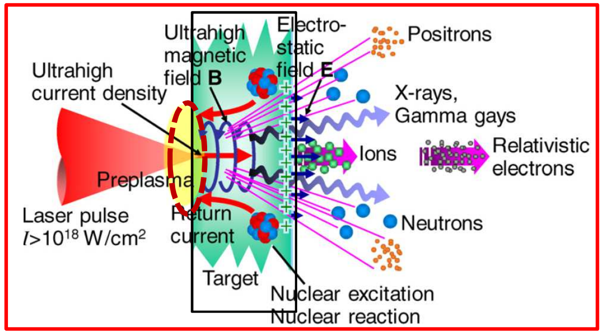
Surface nano-structuring by intense, femtosecond laser. It involves different physics on different time scales in intricated ways → needs appropriate tools to study dynamics



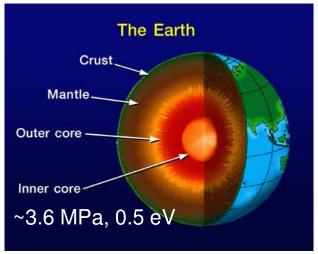


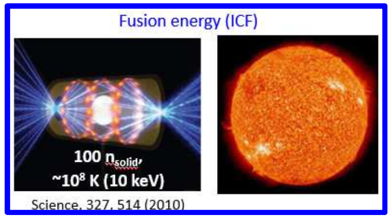
- ~fs (1.) Laser is absorbed by electrons
- fs – ps (2.) Phase transition, plasma creation
- > ps (3.) Thermalization with lattice and ions
- ps – ns (4.) Surface ablation, expansion
- ns - > μs (5.) Re-solidification

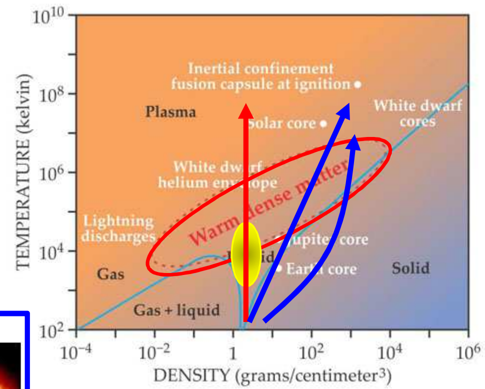
10 – 100 nm depth from the surface



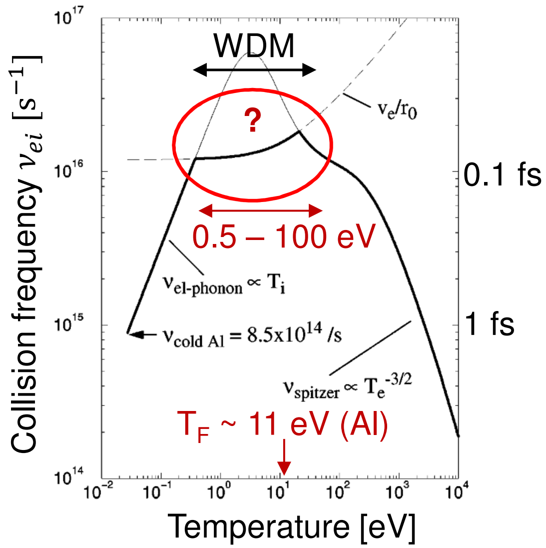
Daido, Nishiuchi and Pirozhkov Rep. Prob. Phys. 75, 056401 (2012)







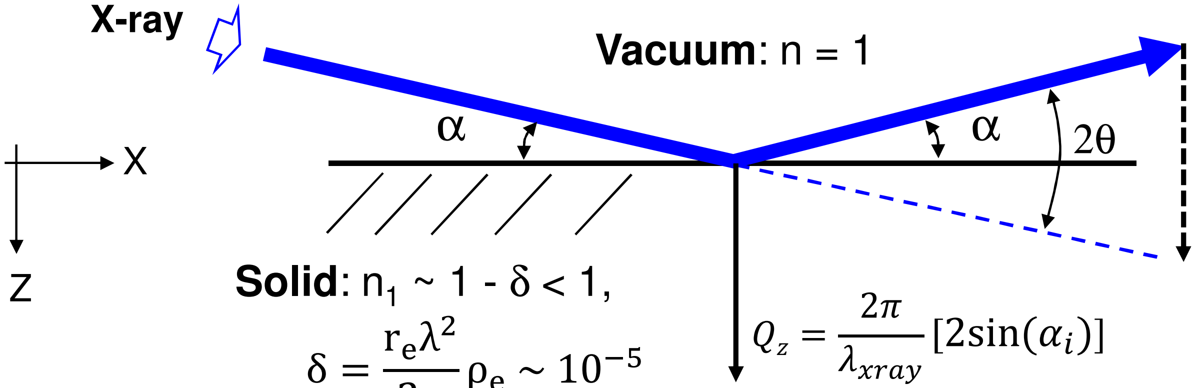
https://physicstoday.scitation.org/do/10.1063/pt.5.7158/full/



➔ Need for *in situ* visualization of surface and subsurface with relevant (Å to sub-μm) resolution with fs – ps precision

First results on "fs" nano-scale surface dynamics upon intense laser irradiation with XFEL + SAXS: T. Kluge et al., PRX **8**, 031068 (2018).

X-ray can be surface sensitive when going to the grazing-incidence (α ≤ 1 deg.)



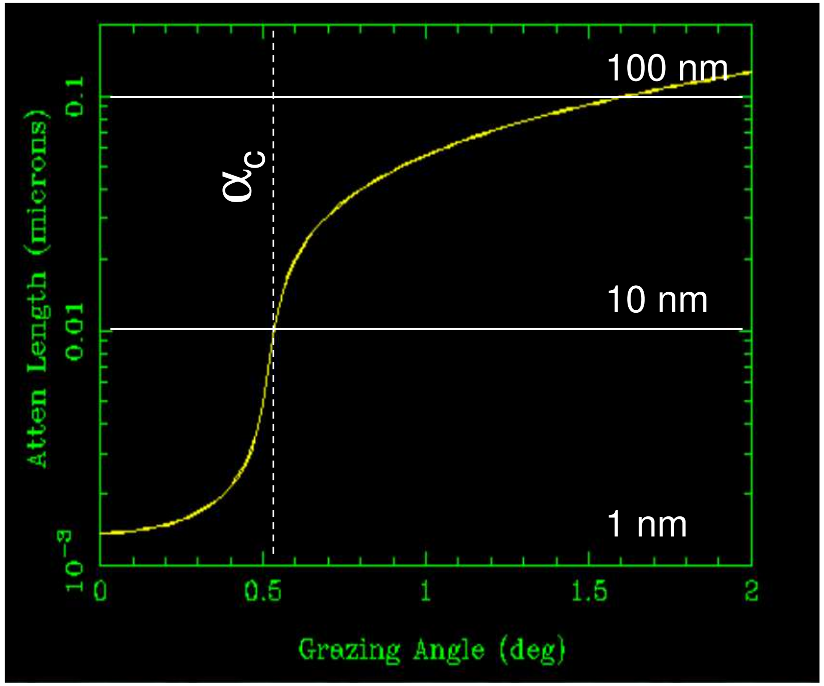
Solid: $n_1 \sim 1 - \delta < 1$,

$$\delta = \frac{r_e \lambda^2}{2\pi} \rho_e \sim 10^{-5}$$

for x-ray

$$Q_z = \frac{2\pi}{\lambda_{xray}} [2\sin(\alpha_i)]$$

Penetration depth for Ta at $h\nu = 8$ keV



http://www.cxro.lbl.gov/

Critical angle (α_c) for total external reflection
for Ta (Z = 73), hν = 8 keV,

$$\alpha_c = \sqrt{4\pi\rho r_0}/k = \mathbf{0.52°}$$

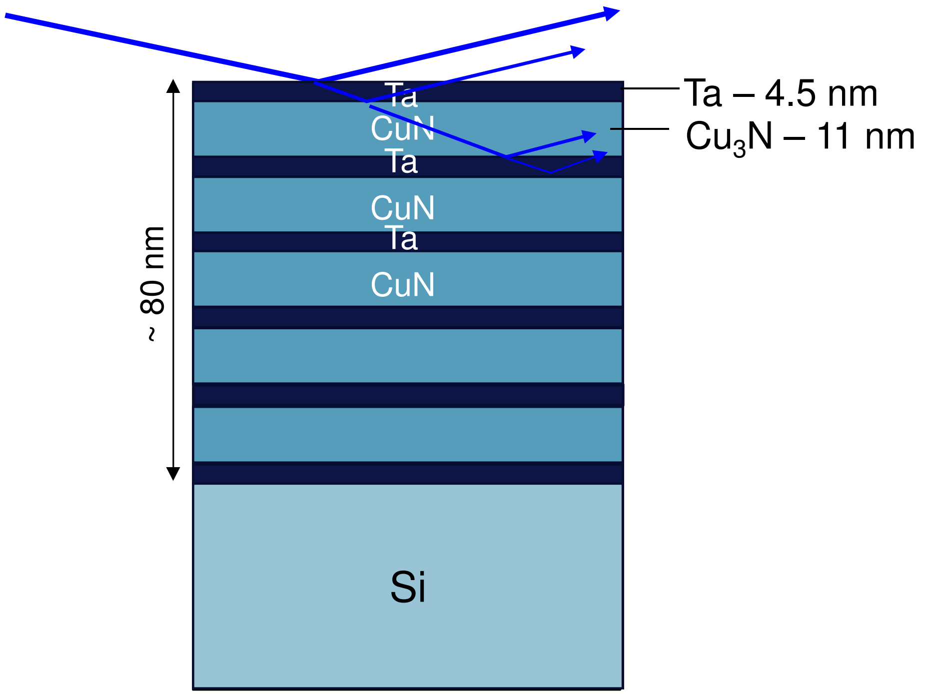
J. Als-Nielsen and D. McMorrow
Elements of modern x-ray physics
John Wiley & Sons, Ltd. (2011)

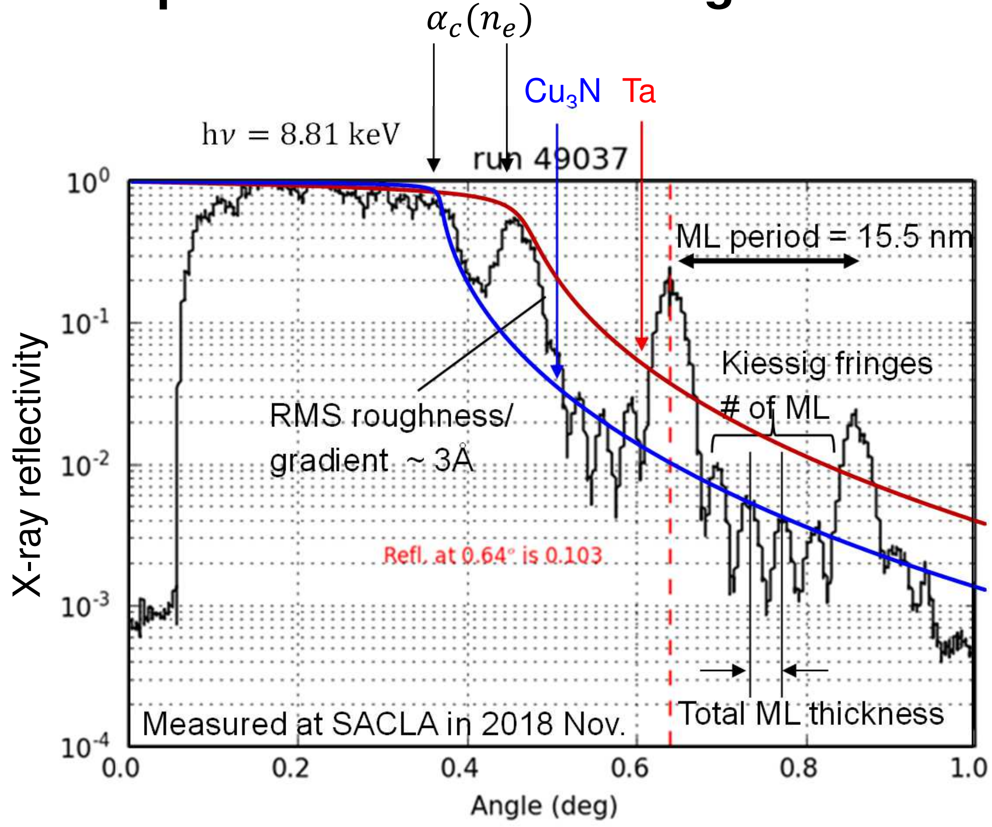
X-ray penetration depth is tunable from a few nanometer (nm) to few 100 nm by changing the grazing-incident angle

➔ **surface sensitive**

X-ray reflectometry (XRR): statistically averaged longitudinal (depth) density profile and roughness properties can be explored – but need a angle scan

Ta / Cu_3N multilayer (ML)
5 repetitions





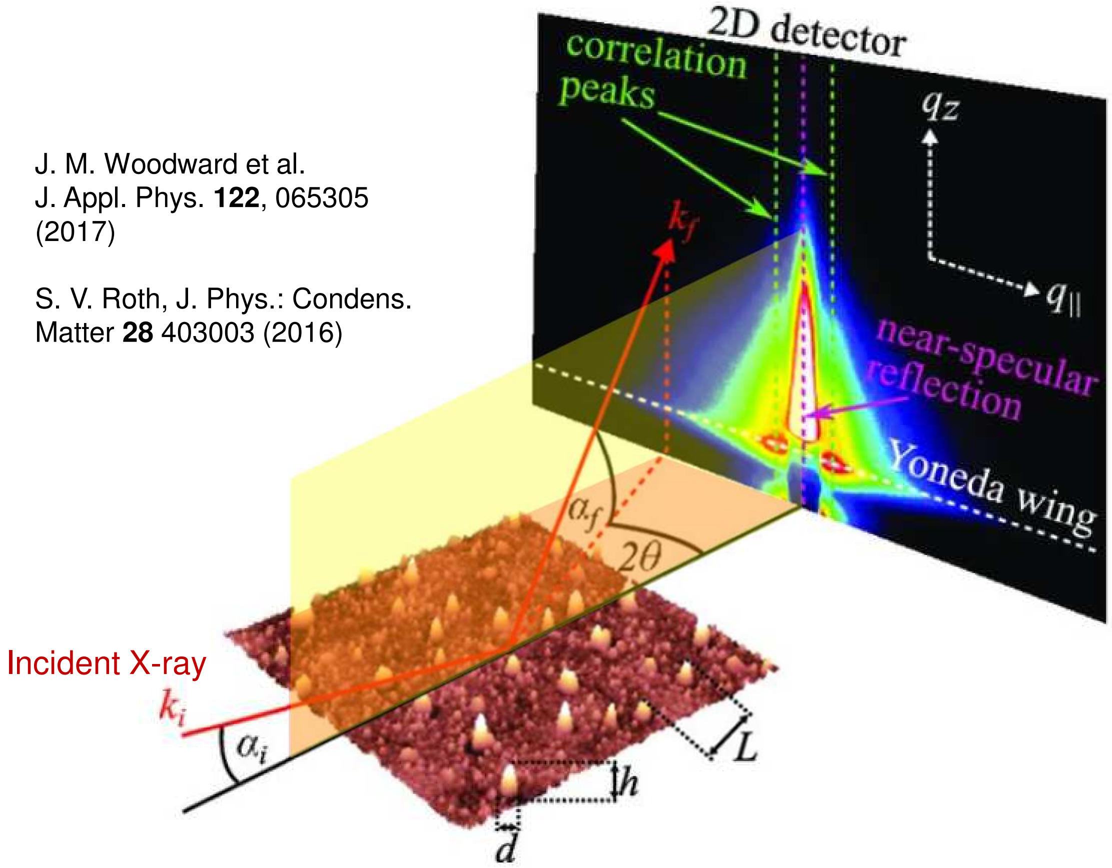
ω - 2θ angle scan

⇨ **Not for a *single shot* experiment**

Grazing-incidence x-ray scattering (GIXS): block the intense specular beam, and look at the weak, diffusely scattered pattern in 2D

J. M. Woodward et al.
J. Appl. Phys. **122**, 065305 (2017)

S. V. Roth, J. Phys.: Condens. Matter **28** 403003 (2016)



GISAXS, in-plane scattering

$$Q_z = \frac{2\pi}{\lambda}\left(\sin\alpha_i + \sin\alpha_f\right) \sim 0.01 \ - \text{few nm}^{-1}$$

→ Information on the longitudinal density profile, RMS roughness

GISAXS, out-of-plane scattering

$$Q_{\parallel} = \frac{2\pi}{\lambda}(\cos\alpha_f \sin 2\theta) \sim 0.01 \ - \text{few nm}^{-1}$$

→ Size and mutual statistical distance of surface nanostructures. Roughness, correlation length, special frequency.

GIWAXS (GID) – large 2θ

$Q \gg 1 \text{ nm}^{-1}$

→ Atomic structure from the x-ray penetration depth ~ 10 – 100 nm

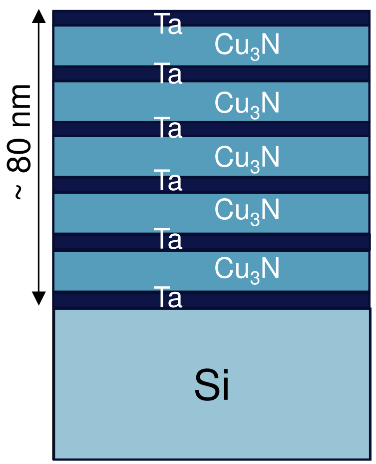
GIXS with a femtosecond intense XFEL for single shot measurement?

First proof-of-principle experiment of grazing-incidence x-ray scattering at SACLA XFEL from multilayers (MLs)

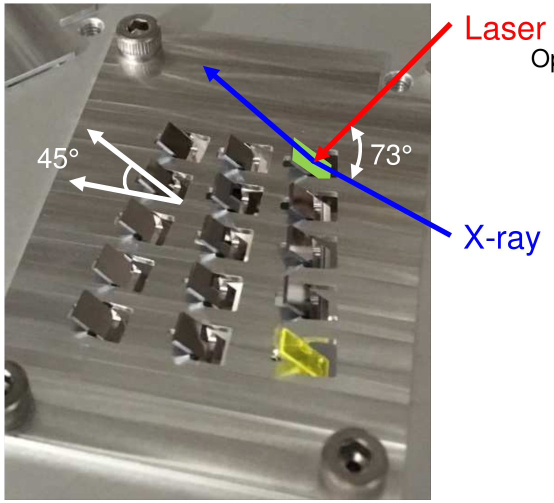
First proof-of-principle experiment was performed at SACLA XFEL facility

- **2 experiments**: Nov. 2018, Feb. 2020
- **X-ray**: ~0.1 mJ, 30 fs, 4 µm spot, **8.81 keV** (to be away from any resonance)
- Station: EH6 at BL3 (Local contact: Toshinori Yabuuchi)
- **Laser**: $10^{14\text{-}15}$ W.cm^{-2} with ~500 µm dia.

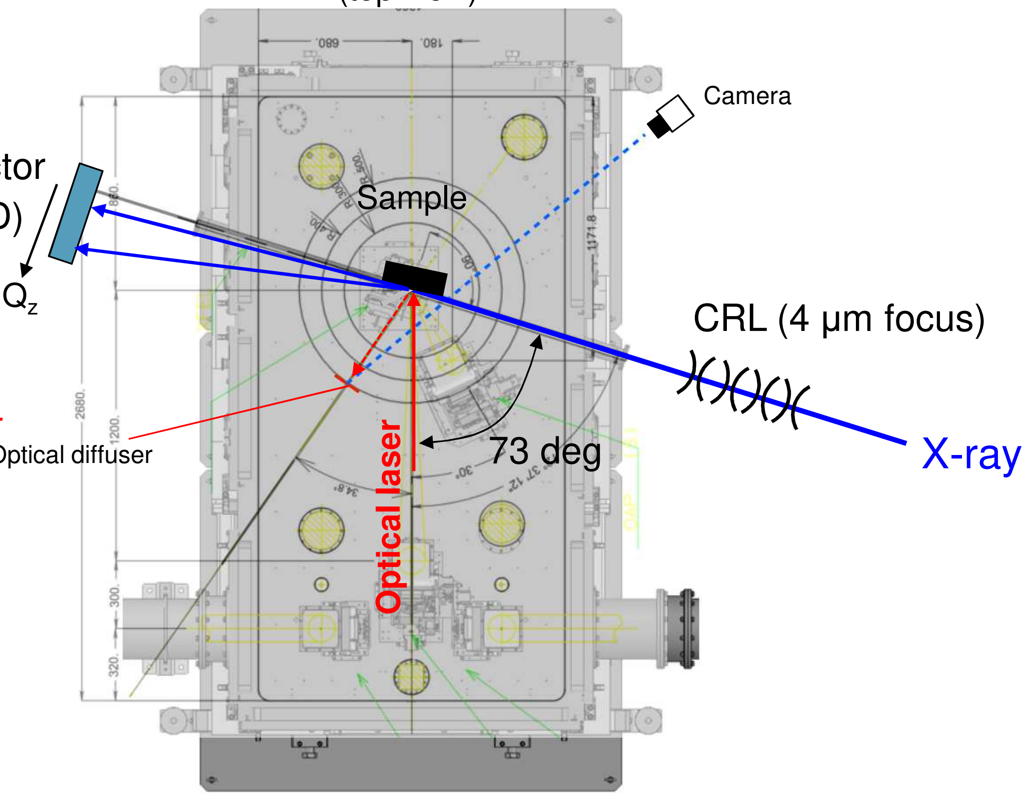
Ta / Cu_3N multilayer (ML)
5 repetitions



Each single-shot sample are isolated



Big vacuum sample chamber (top view)



T. Yabuuchi et al., J. Synch. Rad. 26, 585 (2019).

Specular beam stop

In-plane scattering

$$Q_z = \frac{2\pi}{\lambda}\left(\sin\alpha_i + \sin\alpha_f\right) \sim 0.01 \; - \text{few nm}^{-1}$$

→ Information on depth profile

Out-of-plane ... along Q_y

$$\ldots \alpha_f \sin 2\theta) \sim 0.01 \; - \text{few nm}^{-1}$$

→ Roughness, correlation length, special frequency of roughness (Hurst parameter)

Yoneda peaks

→ Density of the topmost (~few nm) layer

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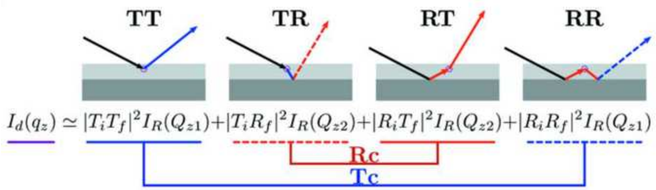
Retrieval of the real-space density profile from BornAgain

Grazing incidence →

~~Kinematical (Born) approximation~~

⇩

Distorted wave Born approximation **(DWBA)**



J. Liu and K. Yager, IUCrJ, **5**, 737 (2018)

L. Randolph, PhD thesis: http://dx.doi.org/10.25819/ubsi/6596

Electron density

Roughness

Diffuse scattering signal

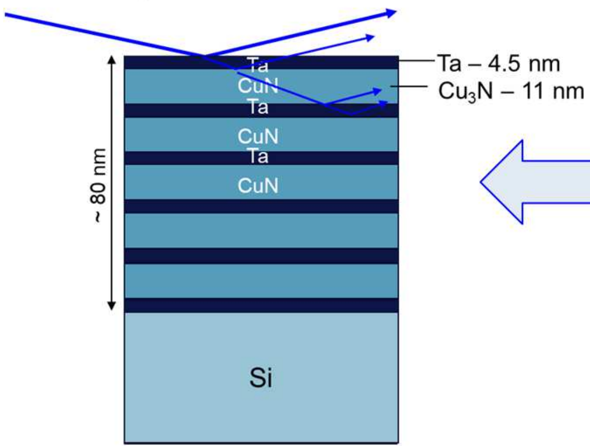
$$I_{diff}(\mathbf{Q}) = \frac{\mathcal{G}\mathbf{k}_1^2}{8\pi^2}\sum_{j,k=1}^{N}\left(n_j^2-n_{j+1}^2\right)\left(n_k^2-n_{k+1}^2\right)^* \cdot \sum_{m,n=0}^{3} \bar{G}_j^m \bar{G}_k^{n*} \exp\left[-\frac{1}{2}\left[\left(Q_{z,j}^m\sigma_j\right)^2+\left(Q_{z,k}^{n*}\sigma_k\right)^2\right]\right] S_{jk}^{mn}(\mathbf{Q})$$

With scattering function

Correlation lengths

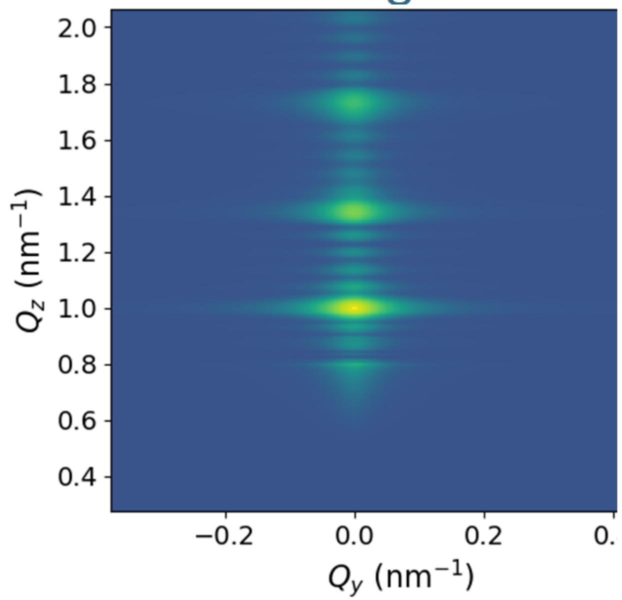
$$S_{jk}^{mn}(\mathbf{Q}) = \frac{1}{Q_{z,j}^m Q_{z,k}^{n*}}\int\left(\exp\left(Q_{z,j}^m Q_{z,k}^{n*} C_{jk}(R)\right)-1\right)\exp(-i\mathbf{Q}_\parallel \mathbf{R})\,d\mathbf{R}$$

Ta / Cu_3N multilayer (ML)
5 repetitions



⇔

BornAgain

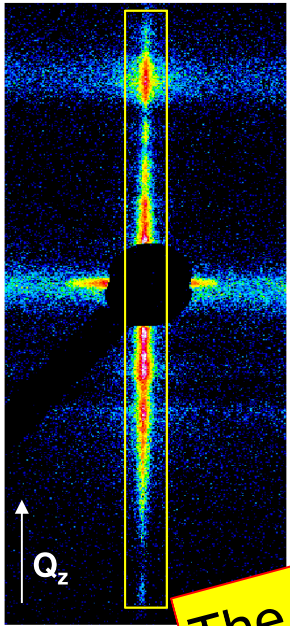


https://www.bornagainproject.org/ G. Pospelov *et al.*, J. Appl. Cryst. **53**, 262 (2020)

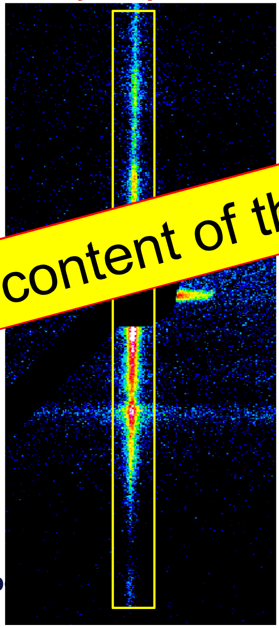
Retrieval of the real-space density profile from BornAgain

Raw scattering data

No laser



With laser
Delay = 3 ps



The content of this slide is removed as it contains unpublished data

L. Randolph, PhD thesis: http://dx.doi.org/10.25819/ubsi/6596

L. Randolph, M. Banjafar et al., arXiv: 2012.15076

Velocity of the density perturbation front agreed with the heat diffusion model

The content of this slide is removed as it contains unpublished data

L. Randolph, M. Banjafar et al., arXiv: 2012.15076

Surface ablation velocity appeared time-dependent

The content of this slide is removed as it contains unpublished data

L. Randolph, M. Banjafar et al., arXiv: 2012.15076

Individual density dynamics does *not* agree with simulations

Experiment

Hydro simulation
(Multi-fs)

Kinetic simulation
(PICLS)

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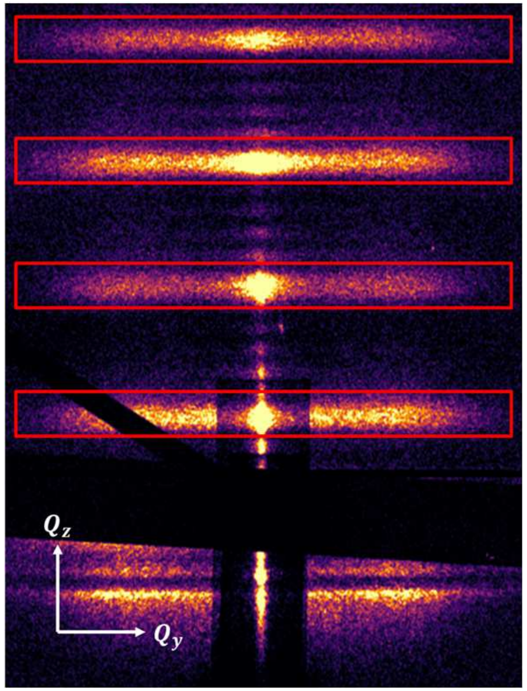
Out-of-plane scattering along Qy gives surface roughness, correlation length, and the Hurst parameter



Diffuse scattering function for self-affine surfaces

$$S_{diff}(Q) = \frac{2\pi}{Q_z^2}\exp(-Q_z^2\sigma^2)\int_0^\infty dR\, RF(Q_z,R)J_0(Q_yR)$$

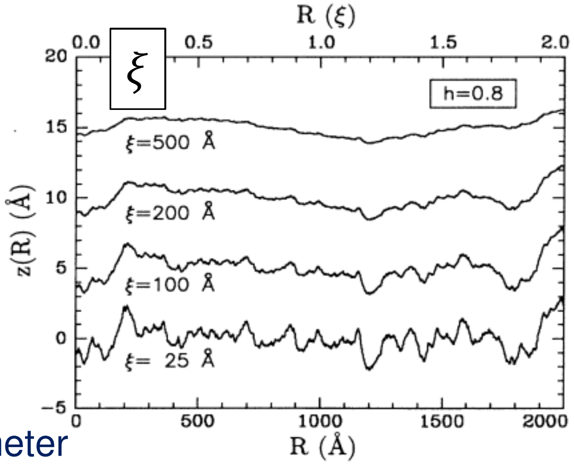
with

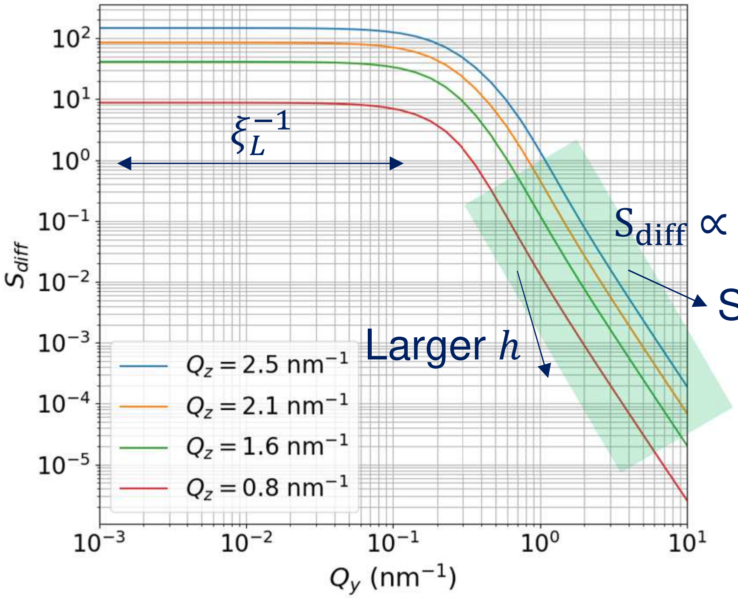
$$F(Q_z,R) = \exp\left(Q_z^2\sigma^2\exp\left(-\left(\frac{R}{\xi_L}\right)^{2h}\right)\right) - 1$$

roughness

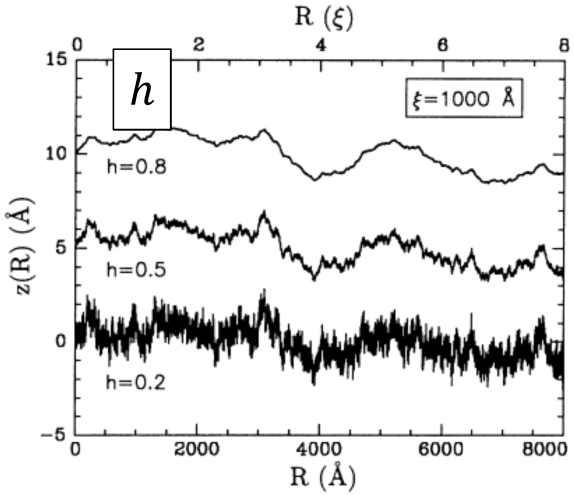
lateral correlation length

Hurst parameter





$S_{\mathrm{diff}} \propto Q_y^{-m};\ m \propto h$



J.-P. Schlomka, et. al. *Phys. Rev. B*, vol. 51, 2311–2321 (1995)

L. Randolph, PhD thesis: http://dx.doi.org/10.25819/ubsi/6596

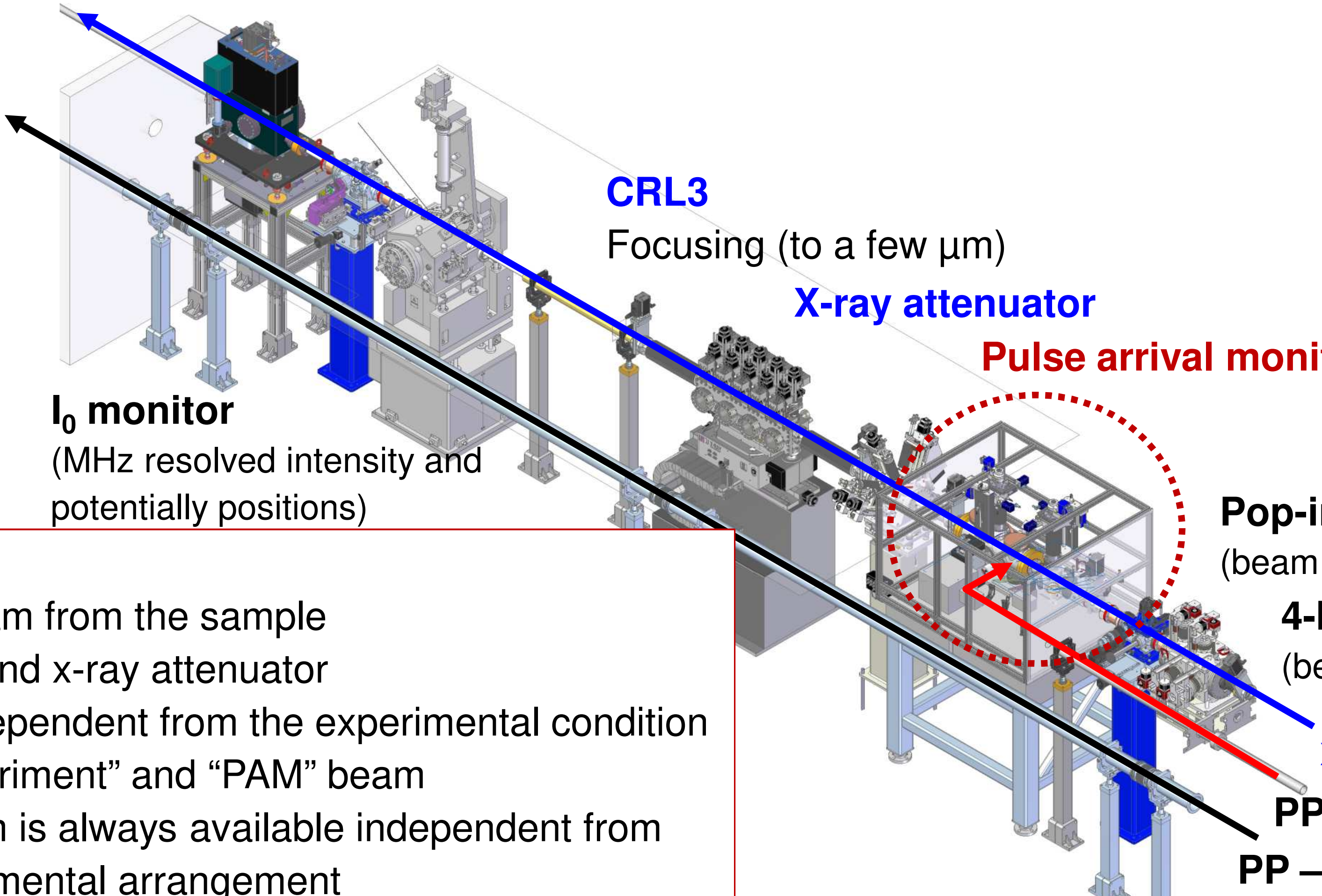
Out-of-plane scattering. Lineout against Q_y contains information on surface structure (roughness, correlation length, special frequency)

The content of this slide is removed as it contains unpublished data

L. Randolph, M. Banjafar, et al., *under preparation*

First PP laser experiment at HED instrument "Surface dynamics and phase transition kinetics of warm-dense-gold" (beamtime #2716, Oct. 2020)

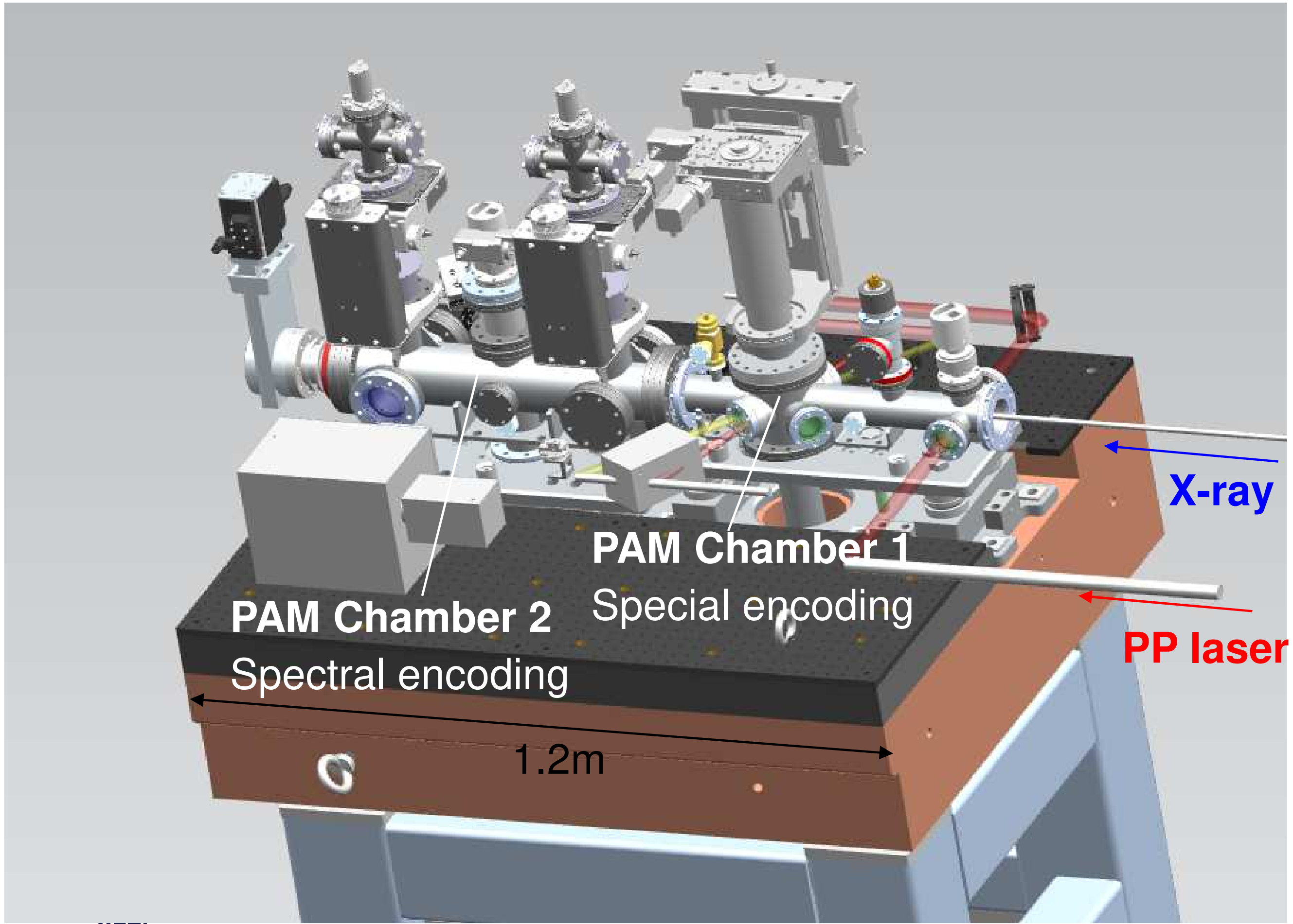
X-ray optics hutch (just before the experimental hutch)



PAM

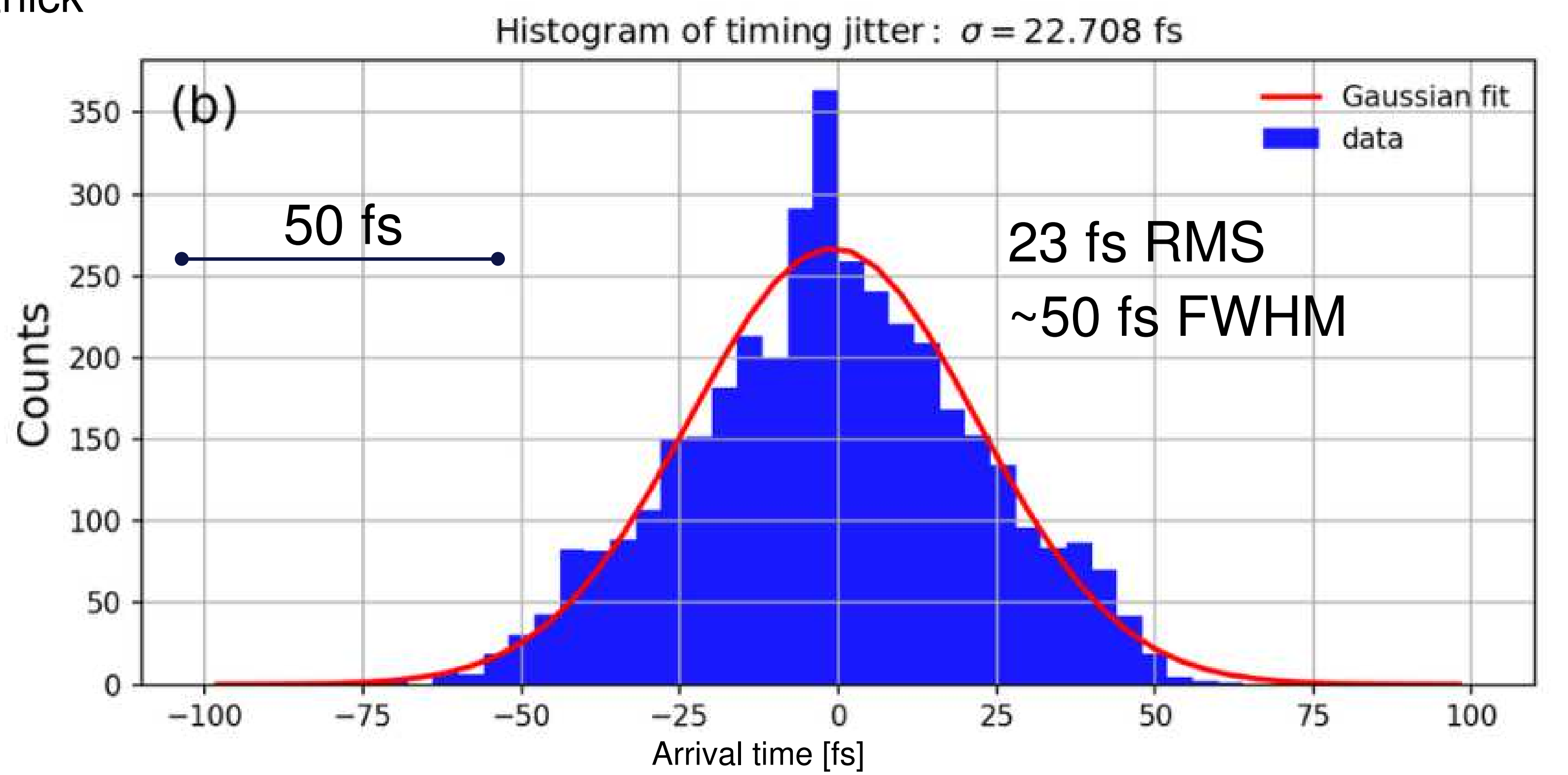
- ~ 10 m upstream from the sample
- Before CRL3 and x-ray attenuator
  - Quasi-independent from the experimental condition
- Split into "experiment" and "PAM" beam
  - PAM beam is always available independent from the experimental arrangement
  - Also works for ReLaX 100 TW laser

2 sample chambers for simultaneous measurement with 2 methods: spatial and spectral encoding.



Arrival jitter measurement between x-ray and PP laser: ~ 23 fs RMS

- Sample: Si_3N_4 2µm thick
- X-ray: 9 keV,
- Transmission to experiment: ~98%



We had a strong contribution from Jia Liu (XPD group, EuXFEL) for the realization of this device and analysis

Simultaneous detection of atomic and nanoscale dynamics at the surface of femtosecond laser-heated warm-dense gold (beamtime #2716 – Oct. 2020)

(PI: J-P. Schwinkendorf / MP: Nakatsutsumi / LC: M. Makita
with main contributions from U. Siegen (L. Randolph, C. Gutt), SLAC (Z. Chen))

- Surface nano-structure dynamics (GISAXS)
- Solid-liquid phase transition (GID / GIWAXS)

M. Z. Mo et al., Science **360**, 1451 (2018);
Z. Chen et al., PRL **121**, 075002 (2018);
V. Recoules et al., PRL **96**, 055503 (2006)

- First PP laser experiment

The content of this slide is removed as it contains unpublished data

Single pulse GISAXS data: major reconfiguration of the surface structure after laser excitation

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Single pulse grazing-incident diffraction (GID / GIWAXS) data

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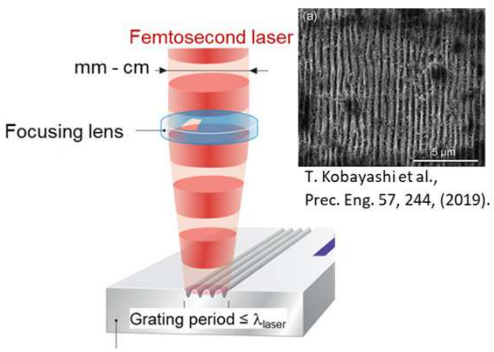
Summary of the 2 experiments (SACLA and EuXFEL) using a grazing-incidence x-ray scattering (GIXS) technique combined with an intense femtosecond laser

- **Using a femtosecond XFEL, one can obtain a GIXS in a single pulse**
  - Track surface and subsurface structure in nano (GISAXS) and Å (GIWAXS) scale *with* high intensity laser
    - ► *Heat transport into bulk*
    - ► *Surface ablation dynamics*
    - ► *Surface nano-morphology dynamics*
    - ► *Phase transition kinetics and change in grain size/orientation*

Outlook

- **GISAXS/GIWAXS with PP laser opens a nice science opportunity in HED field**
  - Phase transition dynamics in laser-heated **warm-dense-matter**.
    - ► Uniform heating = electron ballistic range: 50 – 100 nm = x-ray probing depth.
  - Surface **ablation** dynamics
  - Exploit high repetition rate of the PP laser (100 kHz - 2 mJ, 4.5 MHz - 50 µJ).
    - ► Change in the surface morphology after each laser irradiation (potentially with MHz detectors).
    - ► Change in laser absorption (and subsequent phase transition) due to the surface structure.
    - ► Gives a new insight into *e.g.*, LIPSS dynamics.



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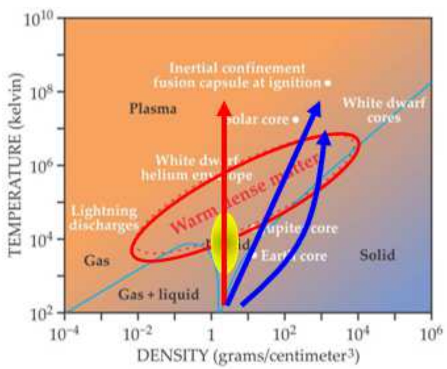

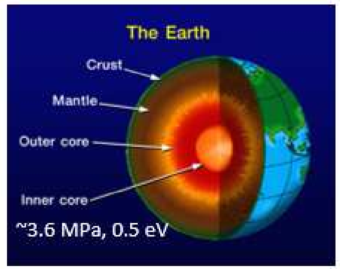

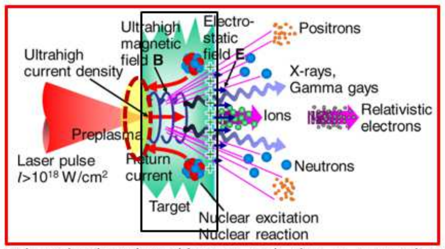

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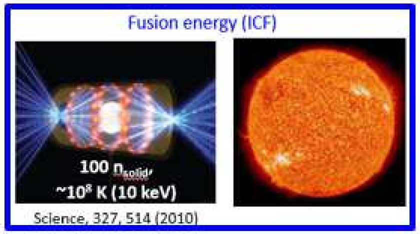

Science, 327, 514 (2010)

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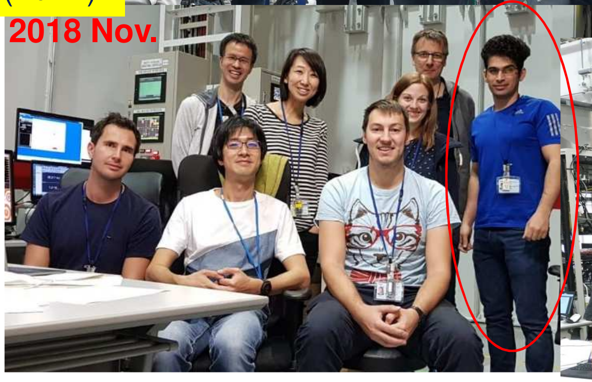

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