Transitions in matter induced by intense X-ray radiation and their diagnostics

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Newly created group at CFEL, DESY:
"X-ray Irradiated Materials: Theory and Computation"

Goal:
computational studies of X-ray irradiated materials relevant for the areas of materials science, diffractive imaging, plasma, and warm dense matter physics investigated with XFEL and synchrotron light sources, with the focus on possible technology development and potential industrial applications.

← FS-CFEL-XM →

Joint initiative of Deutsches Elektronen Synchrotron (DESY) in Hamburg and the Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences (IFJ PAN) in Kraków.

Outline

1. Transitions in matter triggered by X-rays

2. Electronic transitions triggered by low-fluence X-ray pulses

3. Structural transformations at X-ray fluences above damage threshold

4. X-ray induced magnetic transitions

5. Summary

Transitions in matter ...

Energy delivered to a thermodynamic system → transition into a different phase or state of matter

Examples:

Structural transition → leads to a change of a system structure
Magnetic transition → changes magnetic properties (e.g., demagnetization)
Superconductivity → superconducting phase
...

Or

Solid-to-solid → leads to a change of solid's structure
Solid-to-liquid → melting
Solid-to-plasma → ionization
...

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Interaction of X rays with matter:

X-ray photons:

elastic scattering, Compton scattering ← scattering: $\gamma + e_{bound} \rightarrow \gamma'$

photoionization from valence band or core shells, Auger & fluorescence decays of core holes ← ionization: $\gamma + e_{bound} \rightarrow e_{free}$

← Auger decay:

$$h_{core} + e_{bound} \rightarrow e_{free}$$

Electrons:

collisional ionization and recombination from/to bands, thermalization ← electron excitation and relaxation:

$$e_{free} + e_{bound} \leftrightarrow e'_{free} + e_{free}$$

Ions:

charging ← electrostatic repulsion due to Coulomb interaction

$$\leftarrow I^{+n} \quad I^{+m} \rightarrow$$

Main interactions:

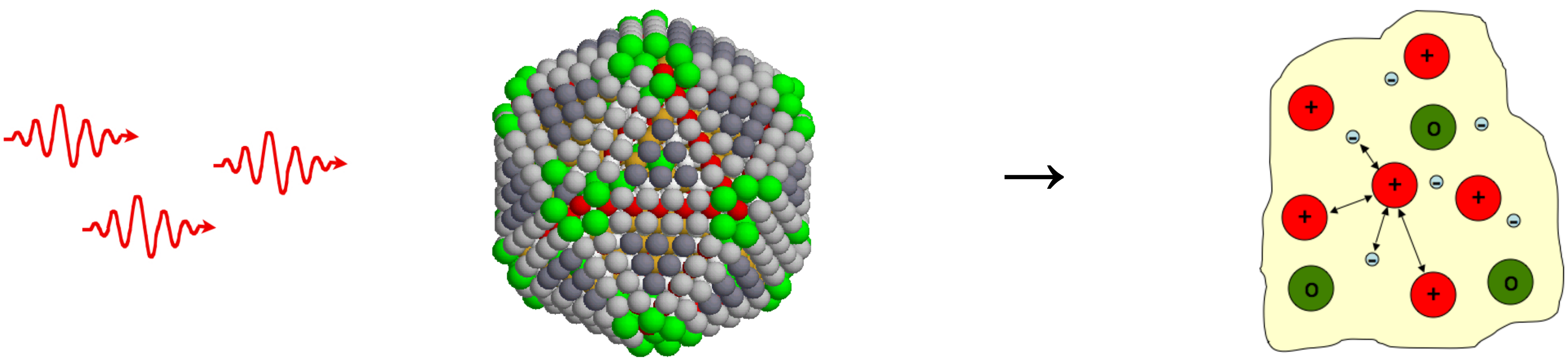
X-ray photons: elastic scattering, Compton scattering, photoionization from valence band or core shells, Auger & fluorescence decays of core holes

Electrons: collisional ionization and recombination from/to bands, thermalization → band modification

→ ELECTRONIC or MAGNETIC TRANSITION

Ions: electrostatic repulsion & band modification →

→ STRUCTURAL TRANSITION



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Transitions in solid materials induced by X-ray radiation

... Femtosecond intense pulses from X-ray free-electron laser ...

FLASH

European XFEL

Transition depends on the average absorbed dose ....

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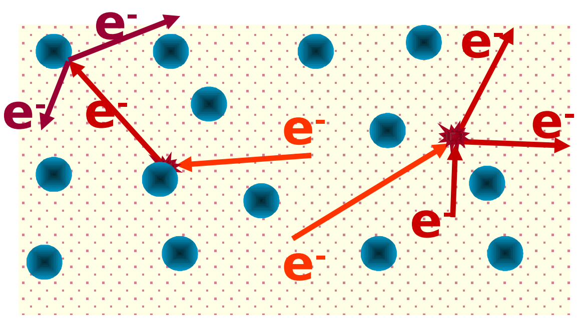
X-ray triggered electronic transitions



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Collaborators for this part:

V. Lipp	**N. Medvedev**	**V. Tkachenko**
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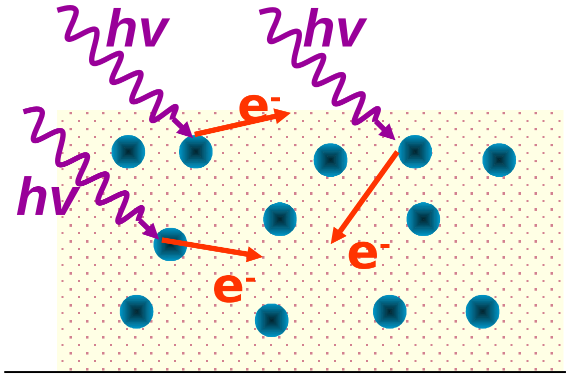
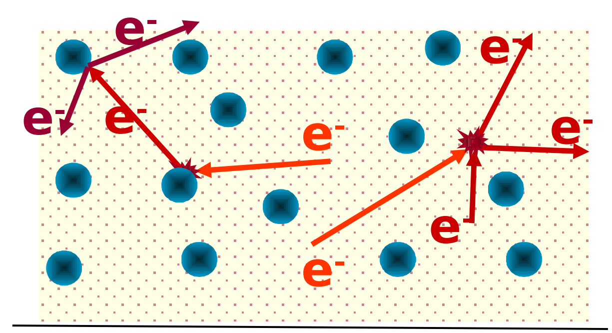
Electronic transitions triggered by low fluence X-ray pulse

Low dose



<u>Photoabsorption</u>
during X-ray pulse:
photo- and Auger
electron emission



<u>Electron kinetics:</u>
impact ionization,
elastic scatterings,
Auger recombination

→ Electronic density increases in the irradiated sample …

Damage Threshold

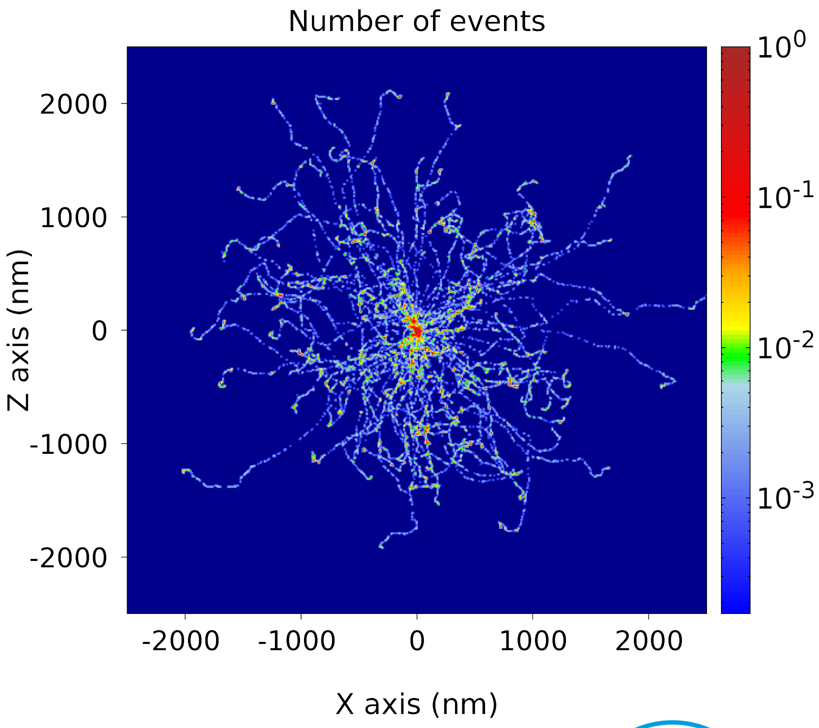
[Images courtesy of N. Medvedev]

Efficient in-house simulation tool: XCASCADE (3D) code

→ Resolves 3D spatio-temporal structure of electron cascades initiated by a primary (photo)electron impact

→ Efficient simulation scheme due to independent electron cascade approximation and low ionization degree of material in which electrons propagate

100 cascades in LiF after 10 keV electron impact



[N. Medvedev, *Appl. Phys. B 118 (2015) 417*]
[V. Lipp, N. Medvedev, B. Ziaja, *SPIE Proc. 10236 (2017) 10236 H*]

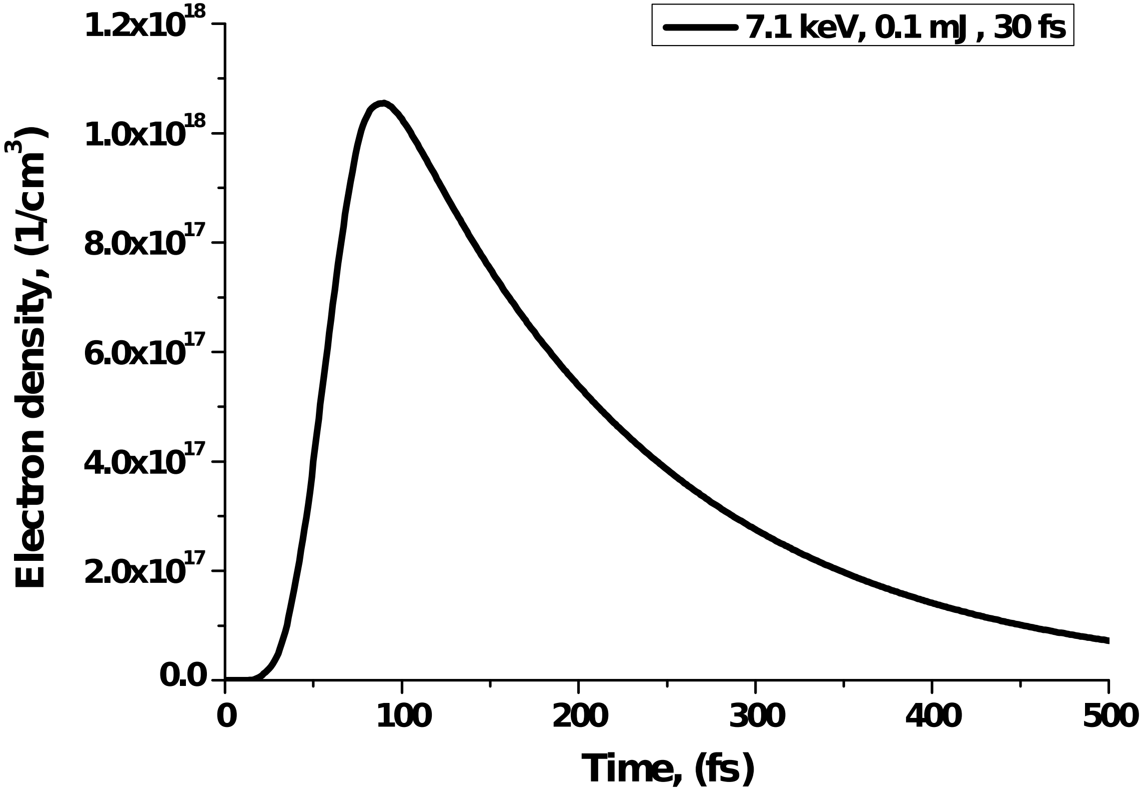
[Images courtesy of V. Lipp]

Electronic transitions triggered by low fluence X-ray pulse

Low dose

Electron density translates into transient change of optical properties

Example:
SiO_2



Damage Threshold

[Images courtesy of N. Medvedev]

Electronic transitions triggered by low fluence X-ray pulse

Low dose

Electron density translates into transient change of optical properties

Example:
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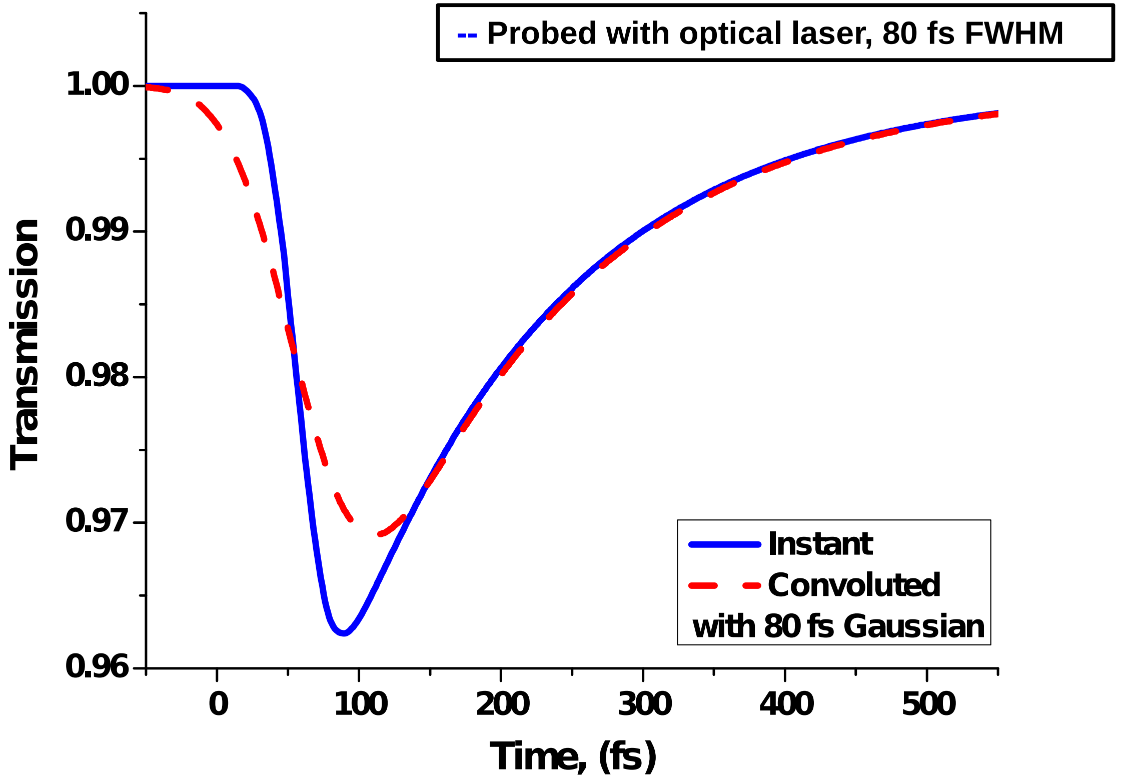
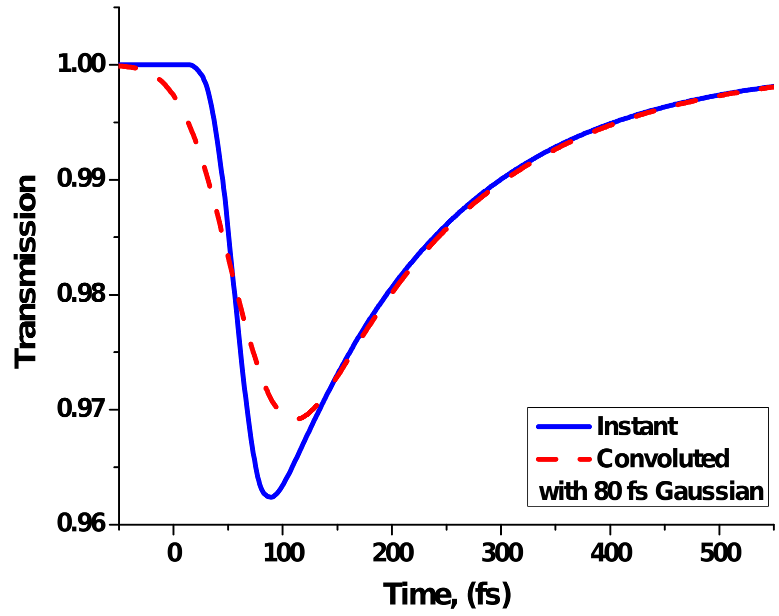


Damage Threshold

Electron density increase triggered by low fluence X-ray pulse

Low fluence Electron density translates into transient change of optical properties

Example:
SiO_2



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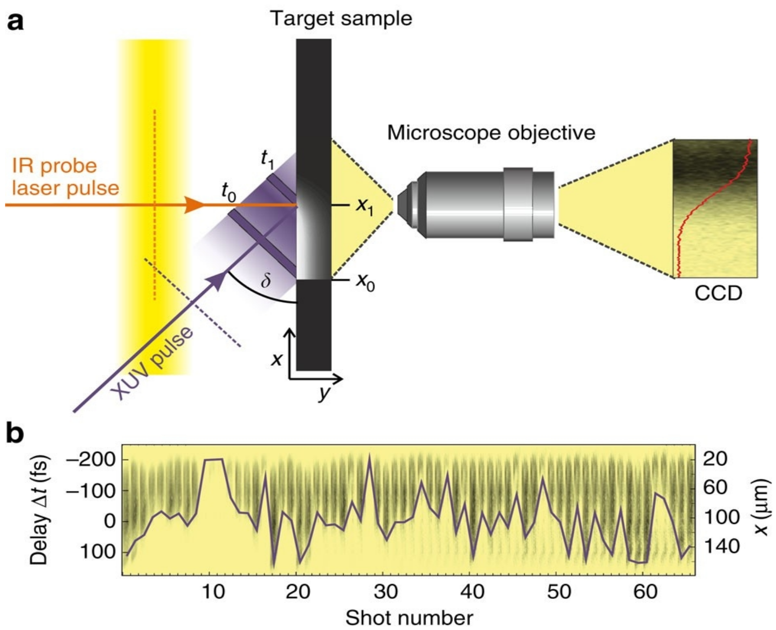


Structural damage fluence

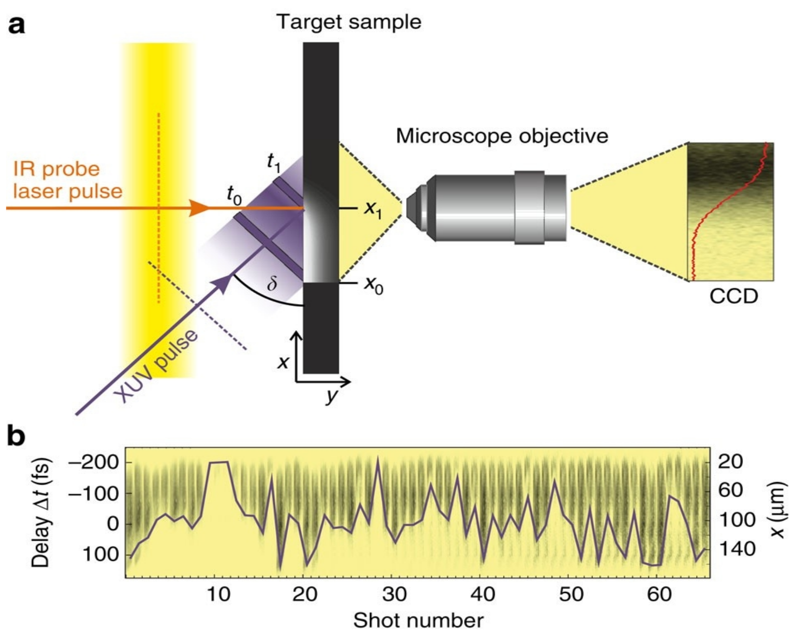
→ Applications for **FEL pulse duration and pulse arrival diagnostics tools**

Electron density increase triggered by low fluence X-ray pulse

Low dose

Electron density translates into transient change of optical properties

→ Applications for FEL pulse duration and pulse arrival diagnostics tools:
[Harmand et al., Nat. Phot. 7 (2013) 215]
[Riedel et al, Nat. Commun. 4 (2013) 1731]
[Finetti et al., PRX 7 (2017) 021043]
[Tkachenko et al., Opt. Lett. 45(1), 33 - 36 (2020)]
[Tkachenko et al., Sci. Rep. (2021) accepted]
...



Damage Threshold

[Image courtesy of R. Riedel]

Outline

1. **Transitions in matter triggered by X-rays**

2. **Electronic transitions triggered by low-fluence X-ray pulses**

3. **Structural transformations at X-ray fluences above damage threshold**

4. **X-ray induced magnetic transitions**

5. **Summary**

X-ray triggered structural transformations

[Courtesy: DESY]

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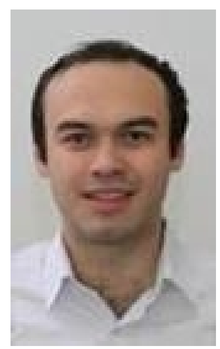

M. Abdullah

DESY

Z. Jurek

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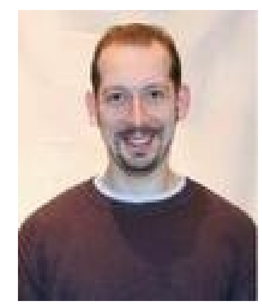

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Structural transformations by X-ray pulses of fluence above the structural damage threshold

Structural damage fluence

Non-thermal transformation (~100 fs) Thermal melting (~ ps)

↑ ↑

Change of interatomic potential induced by presence of many excited electrons

Heating of atomic lattice due to electron-phonon coupling within the same interatomic potential

Melting fluence

Simulations with an in-house hybrid code XTANT combining ab-initio transferable description of band structure (DFTB,TTB) with Parinello-Rahman molecular dynamics for nuclei and Monte-Carlo approach to treat energetic electrons and core holes.

[N. Medvedev, Viktor Tkachenko, V. Lipp, Zheng Li,and B. Ziaja, 4Open 1,3 (2018)]

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[Images courtesy of N. Medvedev]

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Modeling tool: in-house code XTANT using modular hybrid approach

- MD (Parrinello-Rahman scheme) to describe dynamics of nuclei
- MC approach to describe (de)excitation of high energy free electrons in conduction band and creation and relaxation of core holes
- Instanteneous thermalization assumed for electrons within the valence and conduction bands
- Transferable tight binding/DFTB+ module to describe changes of band structure in response to nuclei dislocations; potential energy surface used to derive forces
- Scattering/ionization rates calculated using tables or from complex dielectric function updated at each time step
- Electron-ion coupling treated through Boltzmann collision integral

[Optical pulses: H. Jeschke, M. Garcia, K. Bennemann, PRB 60 (1999) R 3701, PRL 87 (2001) 015003
→ X-rays: Medvedev et al. (BZ): NJP 15 (2013) 015016; PRB 88 (2013) 224304 & 060101; PRB 91 (2015) 054113; PRB 95(2017) 014309]

Diagnostics of transitions in experiments?

Damage thresholds → post mortem measurements on samples

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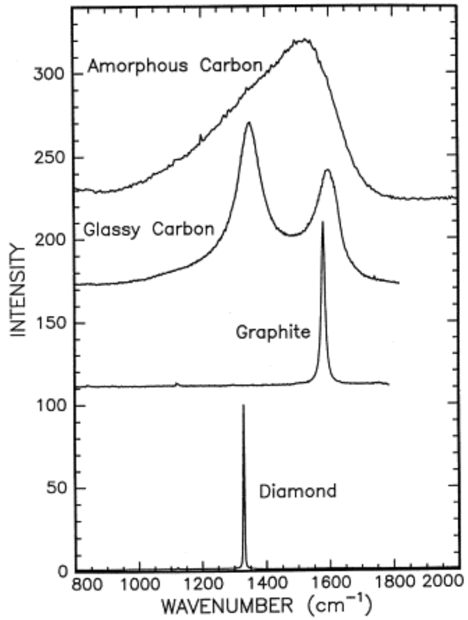


matsci4uwi.wordpress.com

Example: X-ray induced graphitization of diamond

Structural damage fluence

Ultrafast non-thermal process on timescale of 100-200 fs

→ X-ray pulse excites electrons from VB to CB

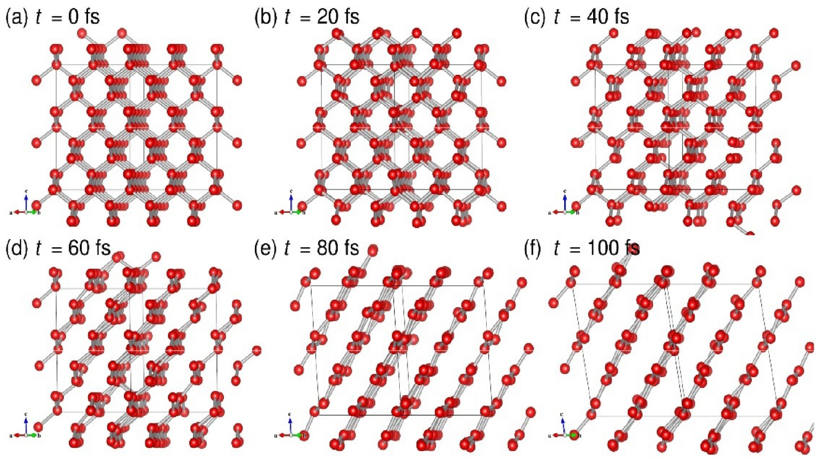
→ the increase of electronic density in conduction band changes interatomic potential; sp^3 → sp^2

→ nuclei rebind to form an (overdense) graphite structure

→ overdense graphite relaxes

Results: Atomic snapshots

Photon energy 92 eV, FWHM = 10 fs



Ultrafast graphitization of diamond

[N. Medvedev, H. Jeschke, B. Ziaja, NJP 15 (2013) 015016]

Melting fluence

Damage thresholds for various X-ray photon energies in good agreement with experiments!

[Medvedev et al. (BZ): NJP 15 (2013) 015016;
PRB 88 (2013) 224304 & 060101;
PRB 91 (2015) 054113]

How to detect transition during experiment?

→ *time-resolved diagnostics of transitions*

Pump-probe experiments:

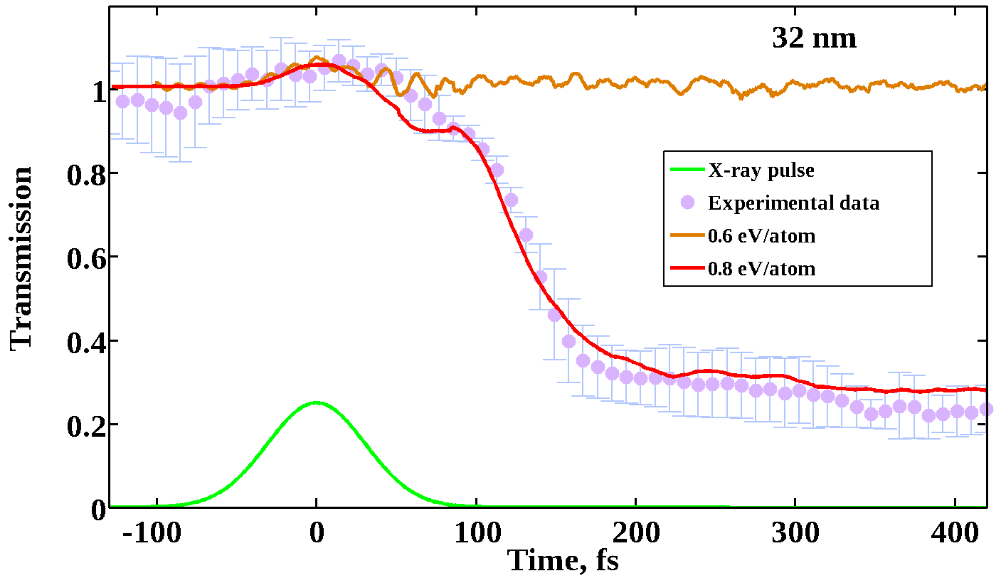
- pump pulse initiates transition ...

- probe pulse probes it at varying time delay …

Transient optical properties as diagnostics of electronic and structural transitions

Damage threshold

Example: First observation of time-resolved graphitization with transmission



Absorbed dose = 0.8 eV/atom

FEL pulse duration = 51 fs

FEL photon energy = 47.4 eV

Probe pulse duration = 32.8 fs

Probe wavelength = 630 nm

X-ray incidence angle = 20°

Experiment performed by Sven Toleikis, Franz Tavella, Hauke Hoeppner, Mark Prandolini et al. at FERMI facility

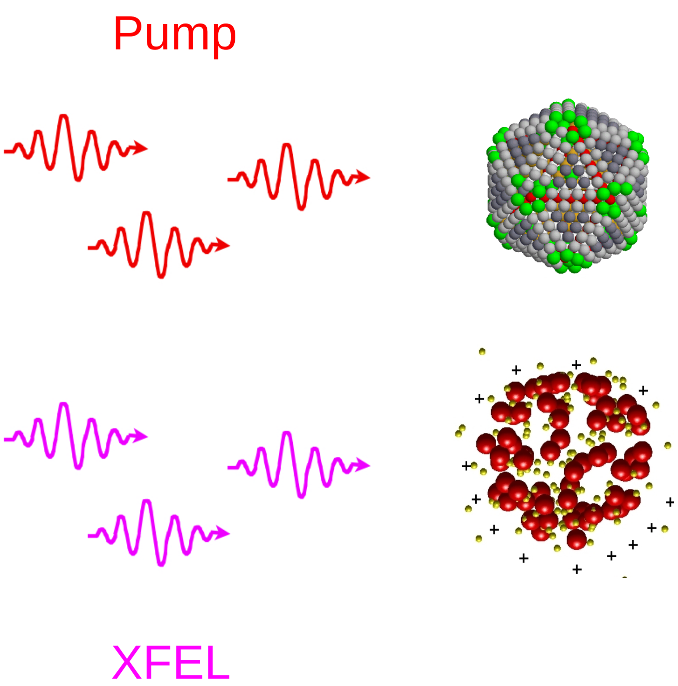
-- Characteristic drop of transmission is observed during the experiment on fs time-scale -- Evidence of phase transition within ~150 fs.

-- Very good agreement with our theoretical predictions for the absorbed dose of 0.8 eV/atom (above graphitization threshold)

Melting threshold

[F. Tavella et al. (V.Tkachenko, N. Medvedev, BZ), HEDP 24 (2017) 22]

X-ray diffraction imaging as diagnostics of structural transitions



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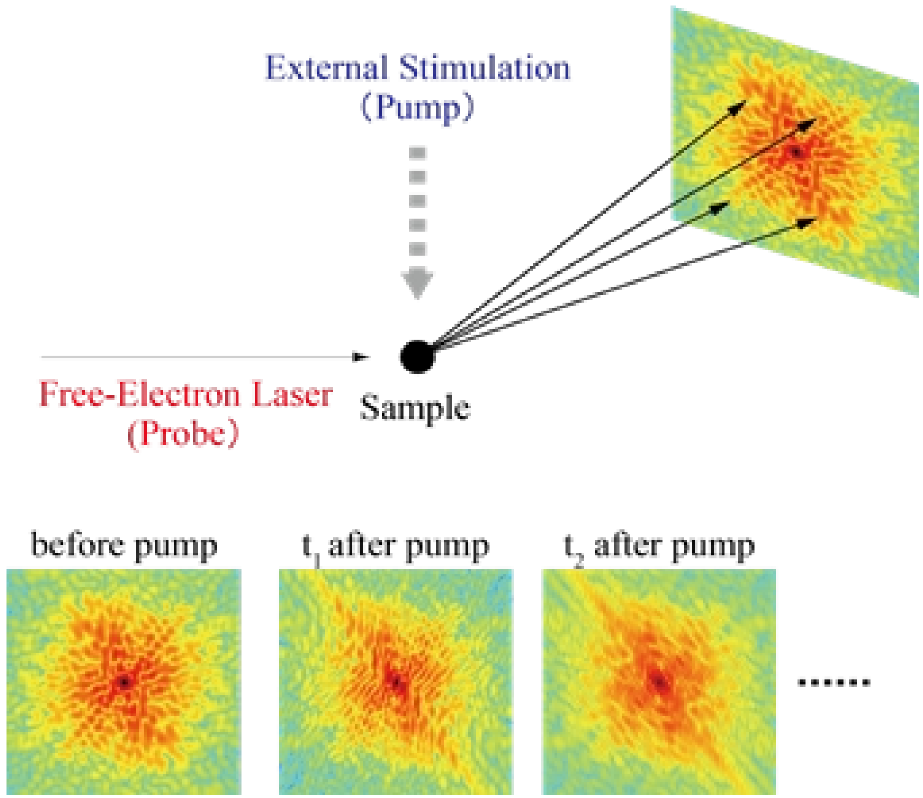


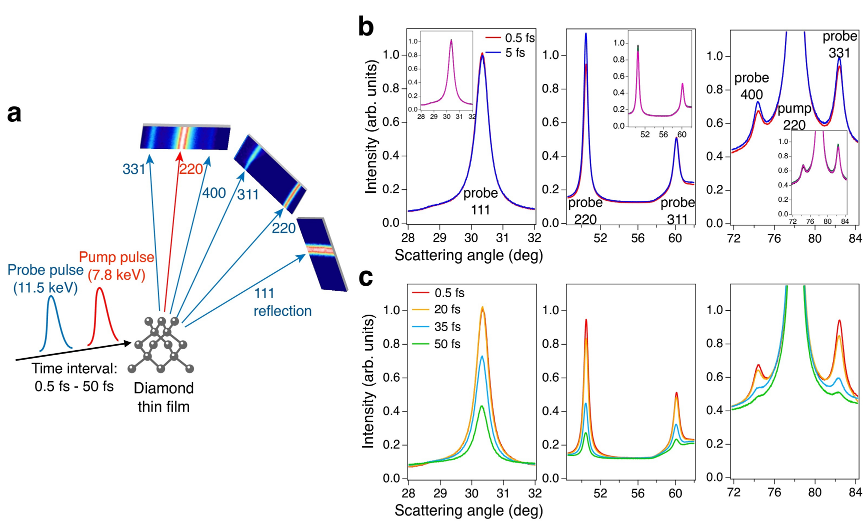
[cxo-www.es.hokudai.ac.jp]

Example: Ultrafast bond breaking in X-ray-excited diamond

Structural damage fluence

Melting fluence

X-ray pump - x-ray probe experiment at SACLA: measured Bragg reflections from diamond film



in various directions (111), (220),(400) etc.

Theory predictions with our in-house code XTANT

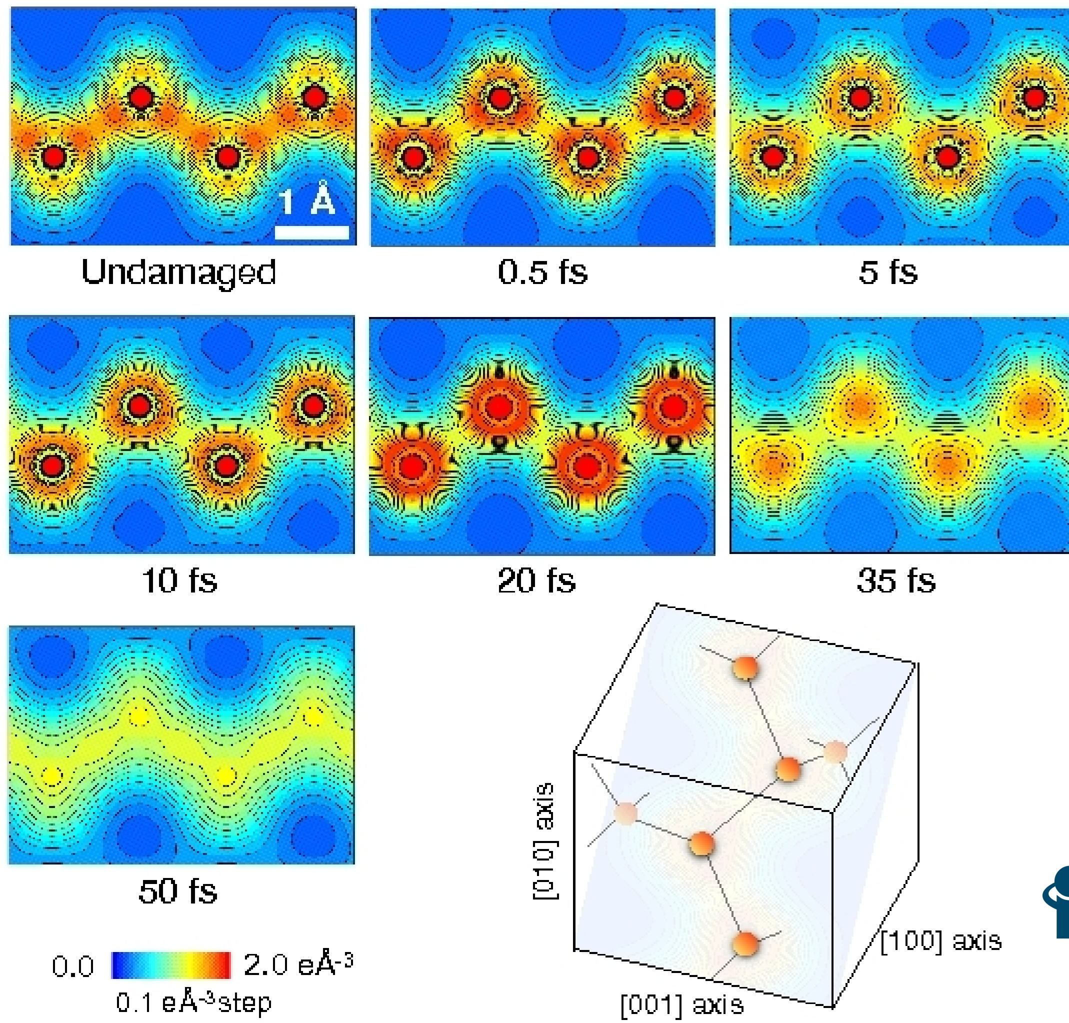
Inoue et al. [PRL 126, 117403 (2021)]

Example: Ultrafast bond breaking in X-ray-excited diamond

Structural damage fluence

Melting fluence

X-ray pump - x-ray probe experiment at SACLA:
valence charge density distribution in (110) plane



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Example: Ultrafast bond breaking in X-ray-excited diamond

Structural damage fluence

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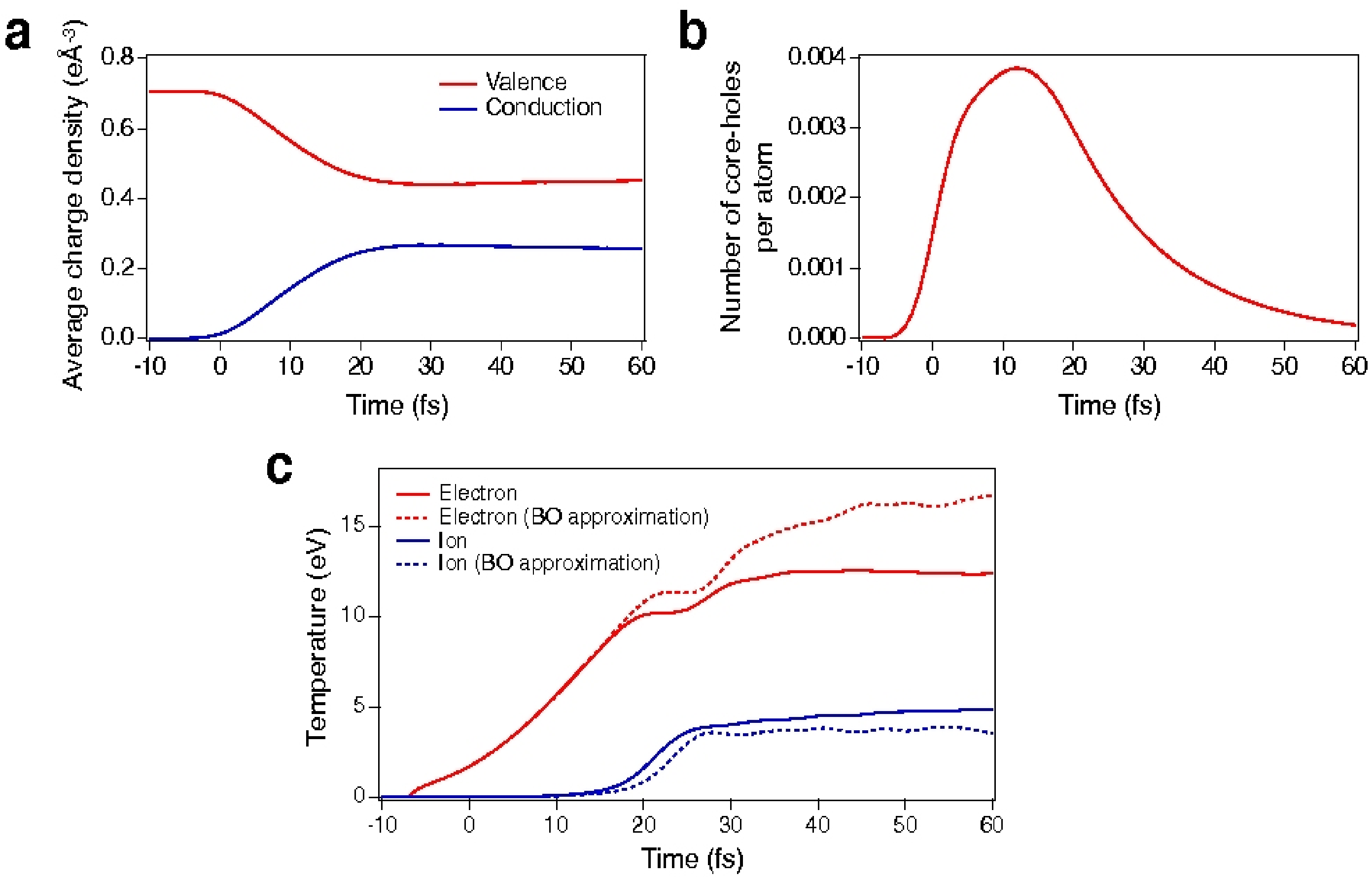

Melting fluence

Theory predictions for (a) average charge density, (b) number of K-holes & (c) electron and ion temperature

→ **ultrafast non-thermal transformation**

Inoue et al. [PRL 126, 117403 (2021)]

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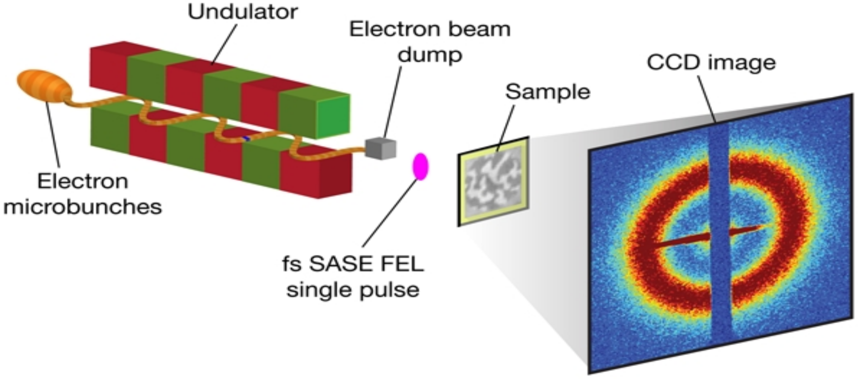
X-ray induced magnetic transitions



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Collaborators for this part:

V. Tkachenko	K. Kapcia	P. Piekarz	A. Kobs	L. Mueller	S. Molodtsov	A. Lichtenstein
IFJ & EuXFEL theory	AMU theory	IFJ theory	DESY exp.	DESY exp.	EuXFEL exp.	EUXFEL/Hamburg U. theory







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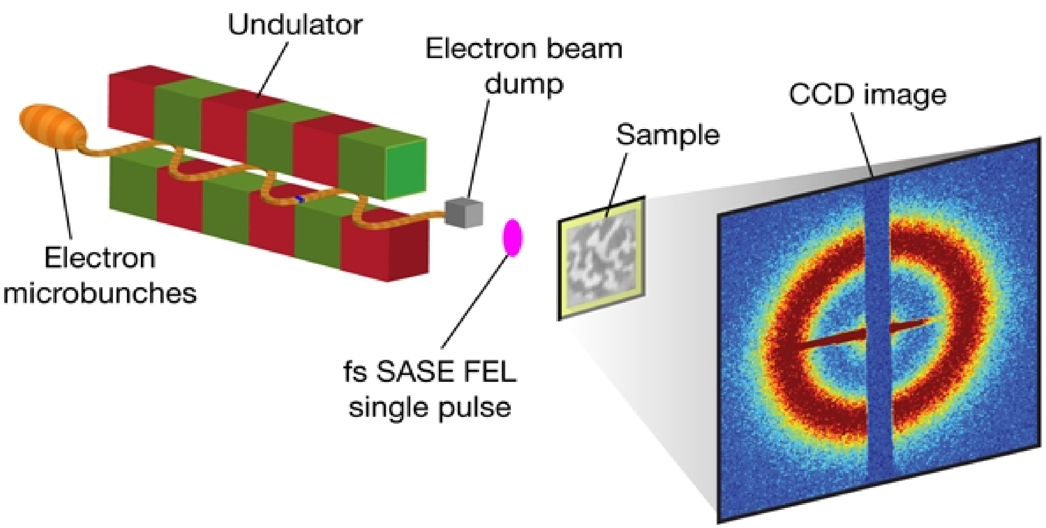
X-ray induced magnetic transitions at low X-ray fluences

Current project status

Experiment performed by A. Kobs, L. Mueller et al. at FERMI on Co M-edge [PRL (2019) under revision]

→ Co/Pt multilayer system irradiated with XUV pulses, 70 fs FWHM of 60 eV photons; mSAXS images recorded
→ normalized scattering strength obtained as a function of pulse fluence
→ scattering efficiency:
$S(Q_{peak}) \sim \int I(t)\, m^2(t)\, dt$
where I(t) pulse intensity and $m^2(t)$ magnetization



[Image courtesy A. Kimel https://physics.aps.org/articles/v3/20]

Preliminary theoretical predictions
→ currently only in the first Co (0.8nm) layer
→ various width Δ of the probed 3d band region tested ← finite width of 3p band

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X-ray induced magnetic transitions at low X-ray fluences

Current project status

Dedicated code XSPIN:

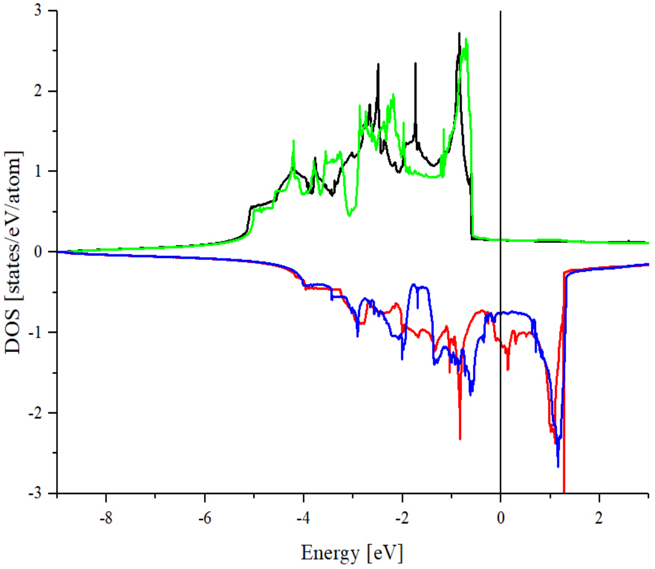
→ uses the description of X-ray irradiation and electronic damage from XTANT

→ introduces two electronic distributions for spin-up and spin-down electrons; spin is conserved in individual electron transitions

→ DOS from DFT (VASP) → electronic energy levels

→ spin flips during the thermalization of total (spin-up + spin-down) electron distribution

→ low-fluence assumption → nuclei kept frozen during the simulation



Comparison between DOS for fcc-Co (bulk lattice parameters, black, red lines) and hcp-Co (bulk lattice parameters, green, blue lines) **[Image courtesy of K. Kapcia]**

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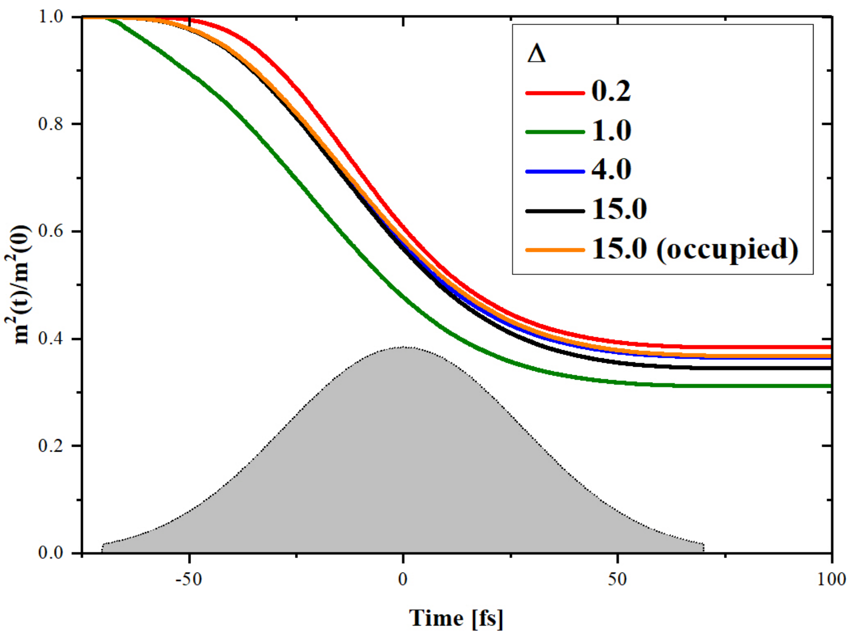
X-ray induced magnetic transitions at low X-ray fluences

Current project status

Preliminary predictions with code XSPIN for Co M-edge:

→ normalized magnetization as a function of time

→ scattering efficiency as a function of pulse fluence will be compared to experimental data



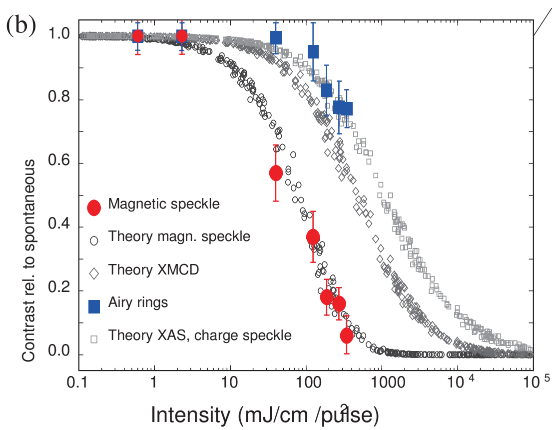
Note: more processes are contributing to dose/fluence conversion in multilayer system!

[Images courtesy of K. Kapcia & V Tkachenko]

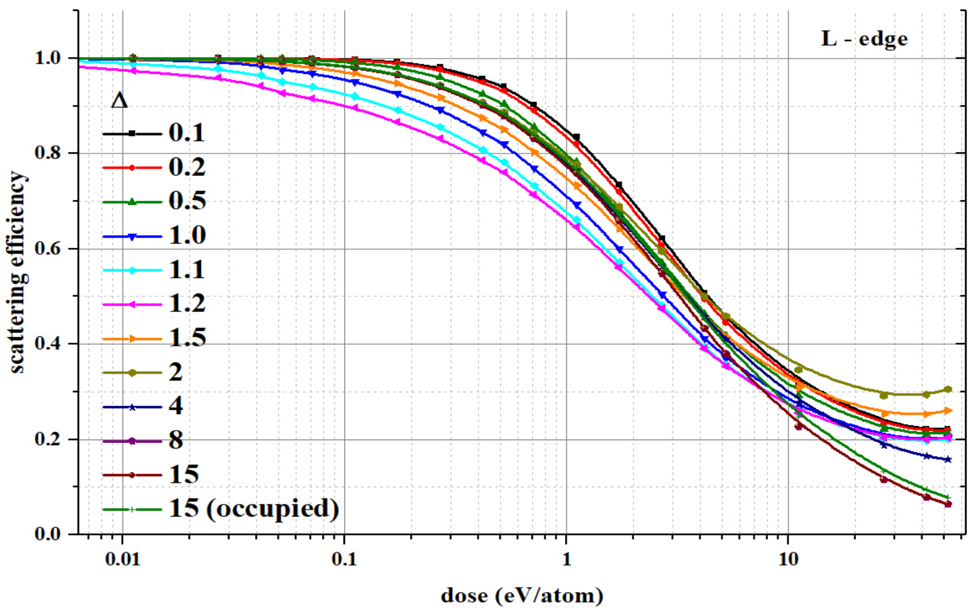
X-ray induced magnetic transitions at low X-ray fluences

Current project status

Preliminary predictions with code XSPIN for Co L-edge:
Experiment by B. Wu et al.[PRL 117 (2016) 027401 for Co/Pd multilayer system
→scattering strength as a function of pulse fluence



[Image courtesy of B. Wu, PRL 117 (2016) 027401]



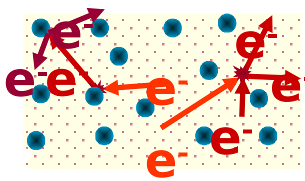
Note: more processes are contributing to dose/fluence conversion in multilayer system!

Further step - complete calculation of scattering efficiency for full multilayer system → on-going

Summary

Transitions in solids induced by X-ray radiation depend on material properties and pulse parameters:

-below damage threshold – non-equilibrium electron kinetics
→ XCASCADE

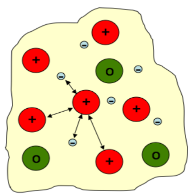


– magnetic transitions → XSPIN

-below melting threshold – also rearrangement of atomic structure:
→ XTANT, XTANT+

-above melting threshold – amorphization; plasma, warm-dense matter formation →XTANT

Diagnostics of transitions:



- transient optical properties ← time-resolved

- X-ray diffraction ← time-resolved

Our computational tools to describe X-ray induced transitions in materials

https://xm.cfel.de/

- **XTANT**: a hybrid simulation tool to study X-ray induced electronic and structural transitions in solids
- **XCASCADE (3D)**: Monte Carlo tool to follow electron cascades induced by low intensity X-ray pulses
- **XSPIN**: a hybrid simulation tool to study X-ray induced magnetic transitions in solids

K. Kapcia

V. Lipp

N. Medvedev

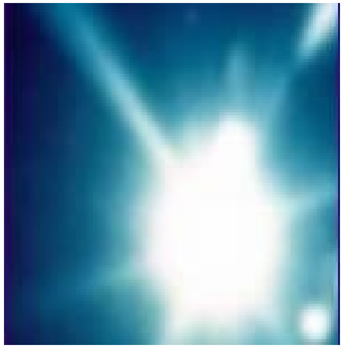
V. Tkachenko

B. Ziaja

Thanking my theory collaborators!

Thanking our external collaborators...

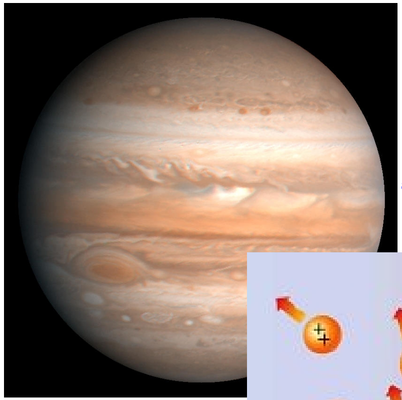
J. Gaudin (CELIA, Bordeaux)



H. Jeschke (U. Frankfurt), Z. Li (LCLS), P. Piekarz (INP, Kraków)

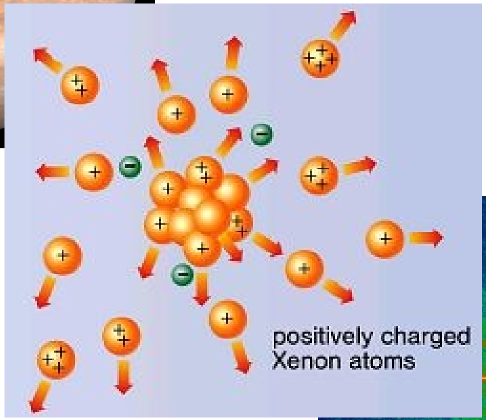
L. Juha, M. Stransky (FZU, Prague), R. Sobierajski (IF PAN, Warszawa)

H.-K. Chung (IAEA, Vienna), R. W. Lee (LBNL, Berkley)



M. Harmand (LULI,CNRS), M. Cammarata (U. Rennes)

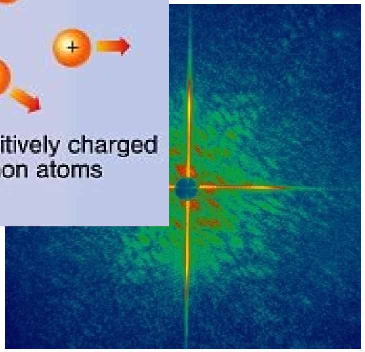
A. Ng (U. British Columbia), Z.Chen, Y.Y. Tsui (U. Alberta), V. Recoules (CEA, DAM)



F. Tavella (LCLS), U. Teubner (U. Oldenburg) and FERMI team

S. Toleikis, H. Hoeppner, M. Prandolini, T. Takanori (DESY)

and ...



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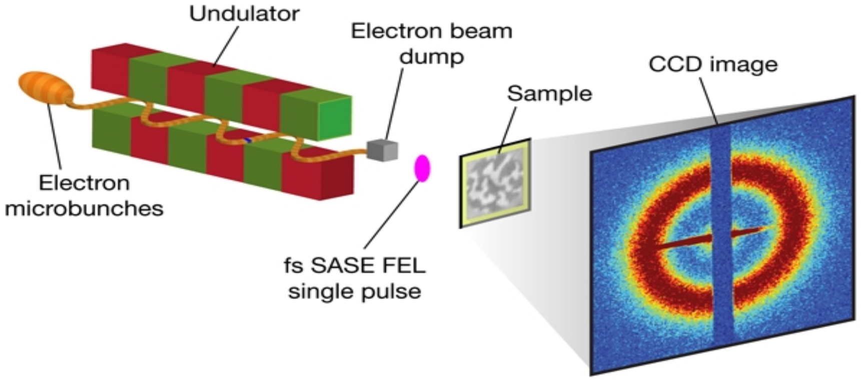
Thanking

European XFEL and IFJ PAN

for

XSPIN project support



[Image courtesy A. Kimel https://physics.aps.org/articles/v3/20]

Thank you for your attention!

CFEL-XM Group

Check also our website: https://xm.cfel.de

Example: X-ray induced ultrafast melting of silicon crystals

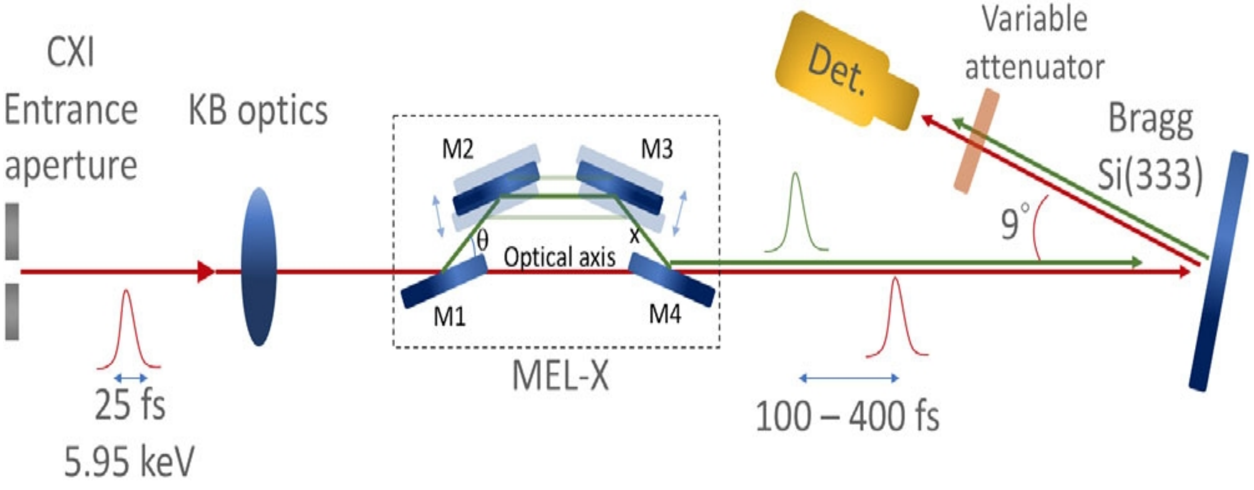
Damage threshold

Melting threshold

Experiment by Pardini et al. at LCLS [PRL 120 (2018) 265701]

single silicon crystal irradiated and probed with hard X-ray pulse;

Bragg reflection Si(333) recorded; 1-2 um^2 focusing



[Image courtesy Pardini et al., PRL 120 (2018) 265701]

Example: X-ray induced ultrafast melting of silicon crystals

Damage threshold

Melting threshold

Predictions with XTANT by Tkachenko et al.

Spatially non-uniform X-ray pulse excites various transitions in Si crystal, depending on the distance from beam focus. Results for 100 %, 50 % and 10 % of nominal fluence are shown →

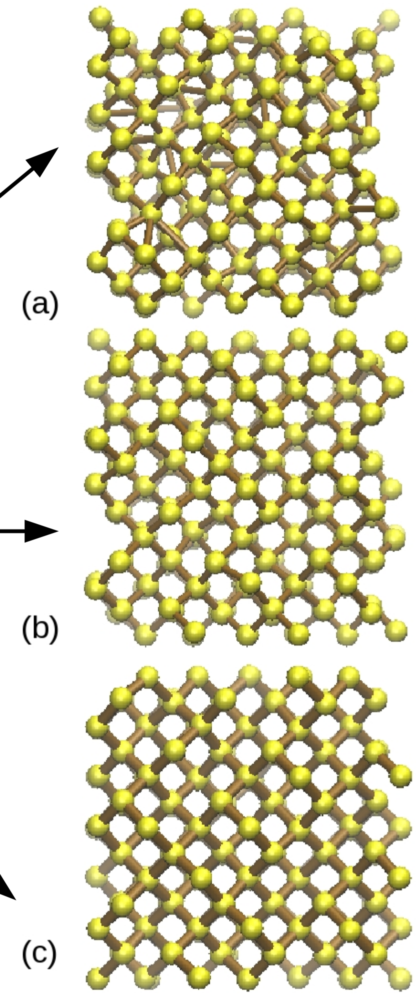
(a) ultrafast non-thermal melting

(b) thermal melting

(c) no structural changes

observed

→ diffracted signal has to be VOLUME INTEGRATED



t=500 fs

Tkachenko et al. [Appl. Sci. 11, 5157 (2021)]

Example: X-ray induced ultrafast melting of silicon crystals

Damage threshold

Melting threshold

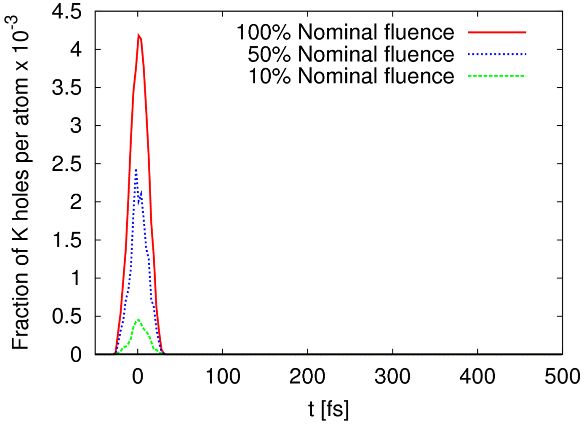
Predictions with XTANT by Tkachenko et al.

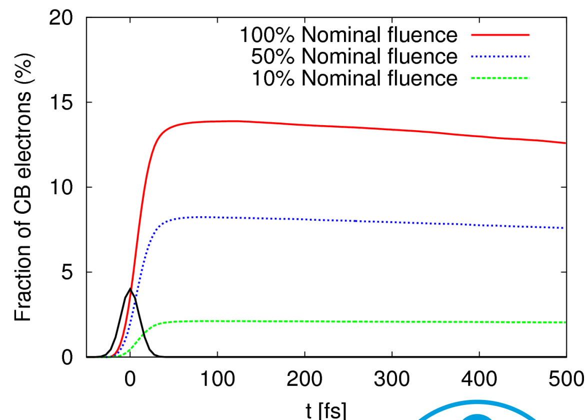
Details on irradiation dynamics depending on the incoming X-ray fluence:

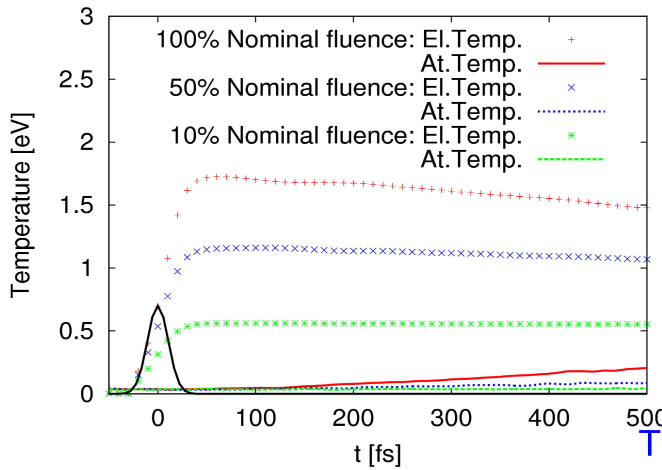
(a) K-hole fraction vs. time

(b) CB electron fraction vs. time

(c) Electron and ion temperature vs time







Tkachenko et al. [Appl. Sci. 11, 5157 (2021)]

Example: X-ray induced ultrafast melting of silicon crystals

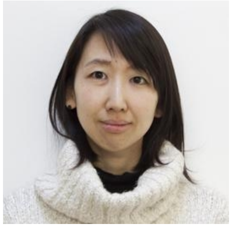
Damage threshold

Predictions with XTANT by Tkachenko et al.

Predicted signal has also to be volume integrated. Integration performed with the code XSINC [M. Abdullah, Z. Jurek, R. Santra, J. Appl. Cryst. 49 (2016) 1048]

M. Abdullah
DESY

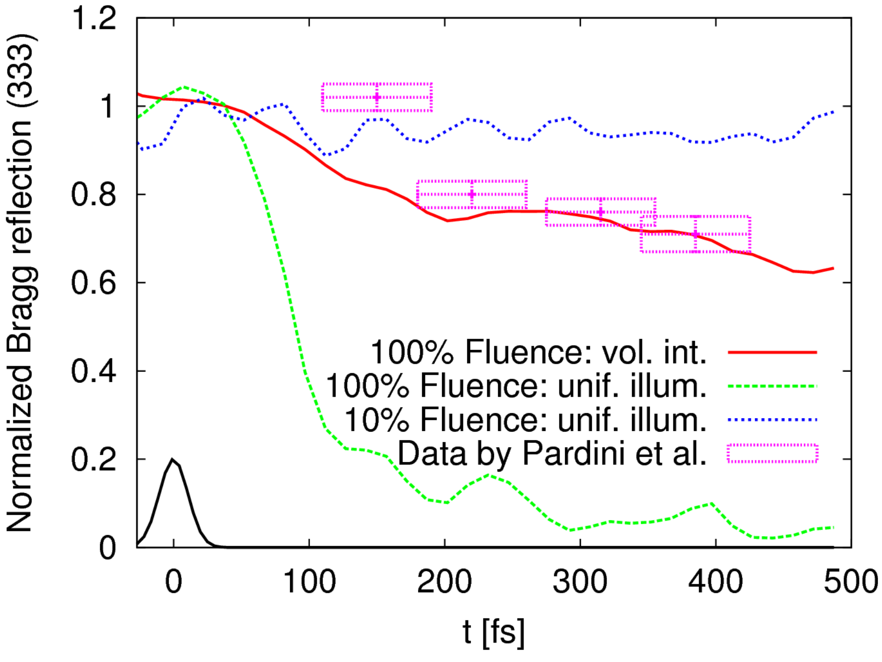
Z. Jurek
CFEL/DESY



M. Makita
EuXFEL

Melting threshold

→ very good agreeement with experiment (note that the data point at 150 fs was excluded in the recent Erratum to exp. paper)



[Pardini et al. PRL 120 (2018) 265701, Erratum PRL 124 (2020) 129903 (E)]

[Tkachenko et al.,Appl. Sci. 11, 5157 (2021)]

Example: X-ray induced ultrafast melting of silicon crystals

Damage threshold

Predictions with XTANT by Tkachenko et al.

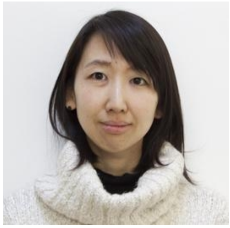
Predicted signal has also to be volume integrated. Integratio

Melting thresho

Sample was non-uniformly illuminated. X rays induced different processes in various crystal regions.

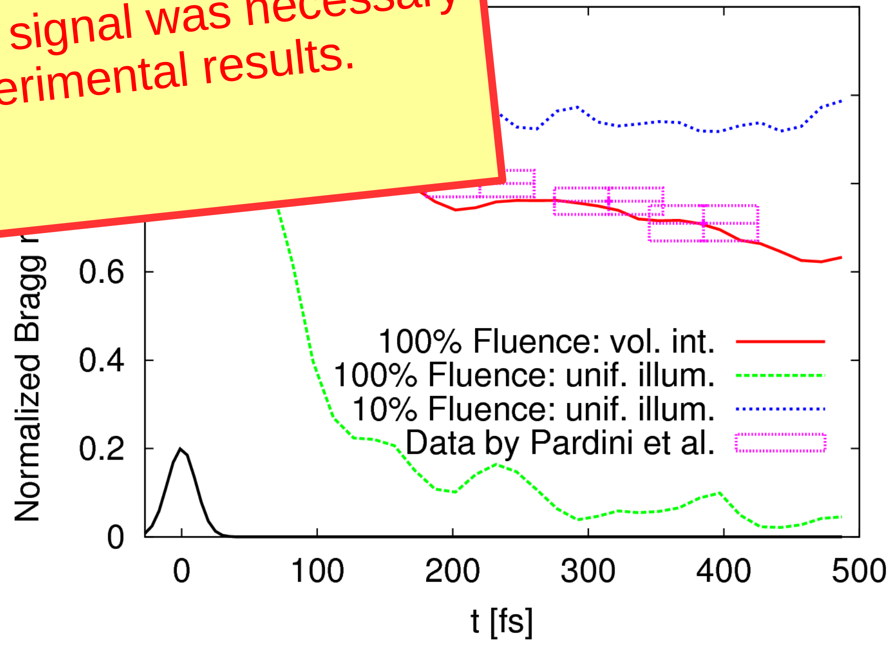
→

Volume integration of the predicted signal was necessary for the correct interpretation of experimental results.

urek
EL/DESY

M. Makita
EuXFEL



[Pardini et al. PRL 120 (2018) 265701, Erratum PRL 124 (2020) 129903 (E)]

[Tkachenko et al.,Appl. Sci. 11, 5157 (2021)]