Modelling the evolution of X-ray free-electron-laser irradiated solids towards warm-dense-matter state

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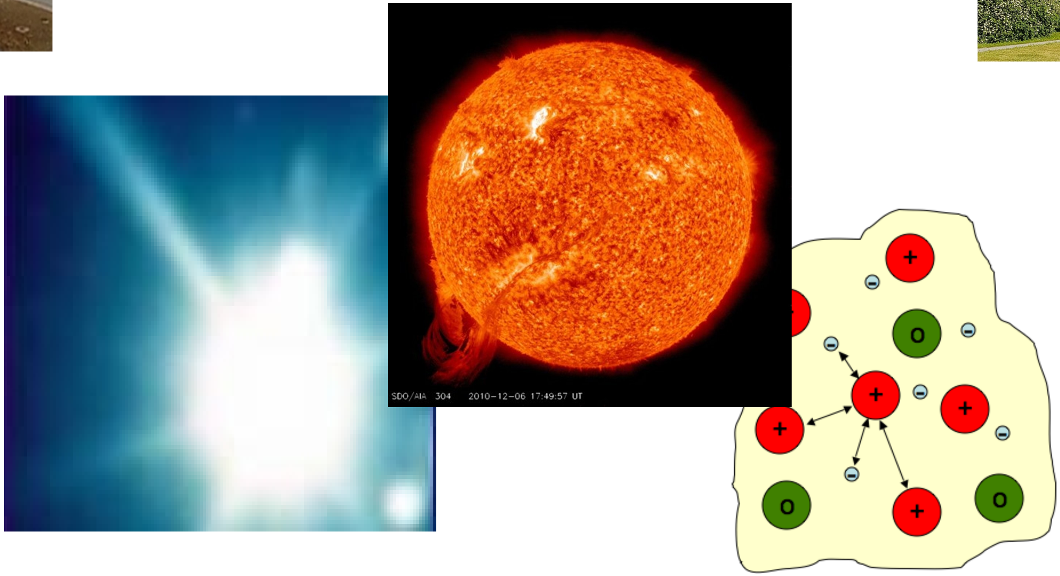


CFEL SCIENCE

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

DESY

Recently 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, Germany and the Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences (IFJ PAN) in Kraków, Poland.

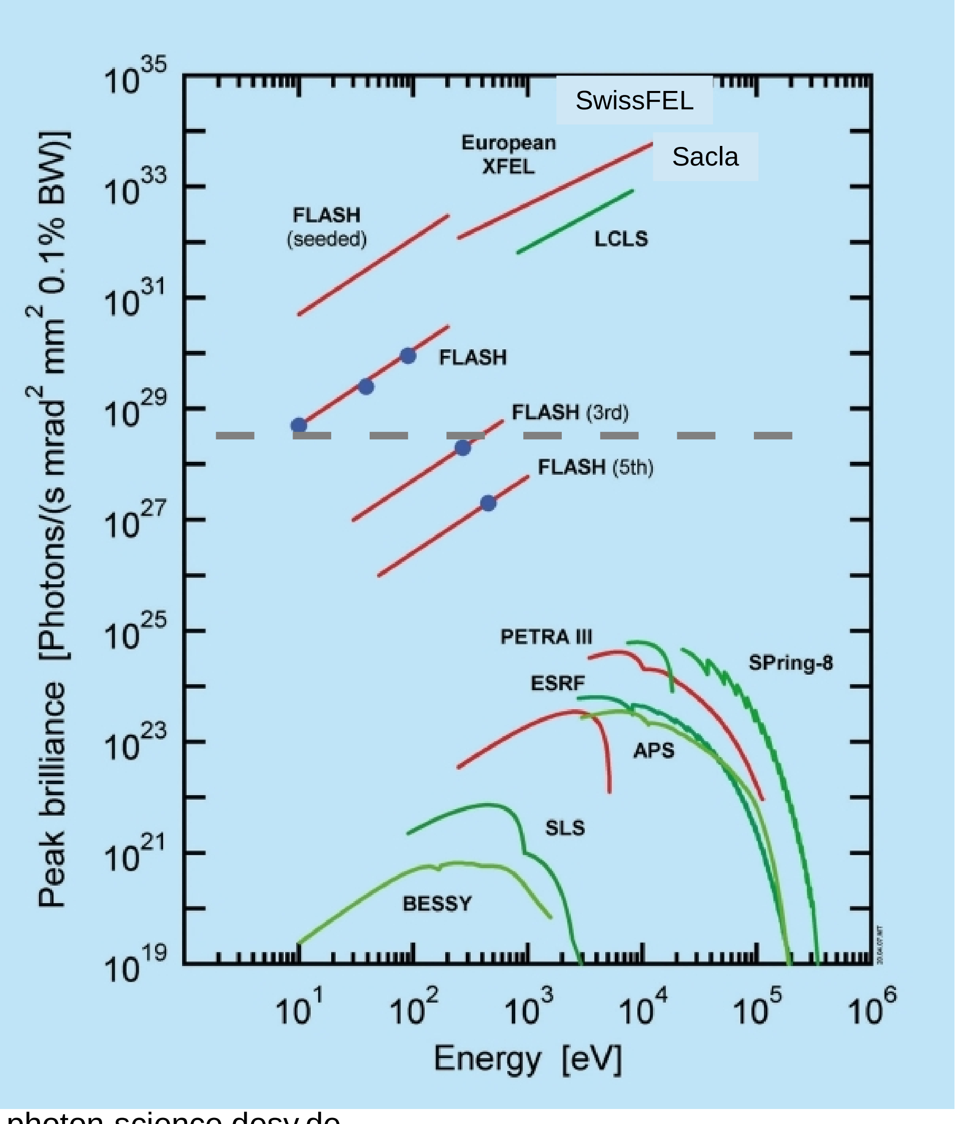
Outline

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

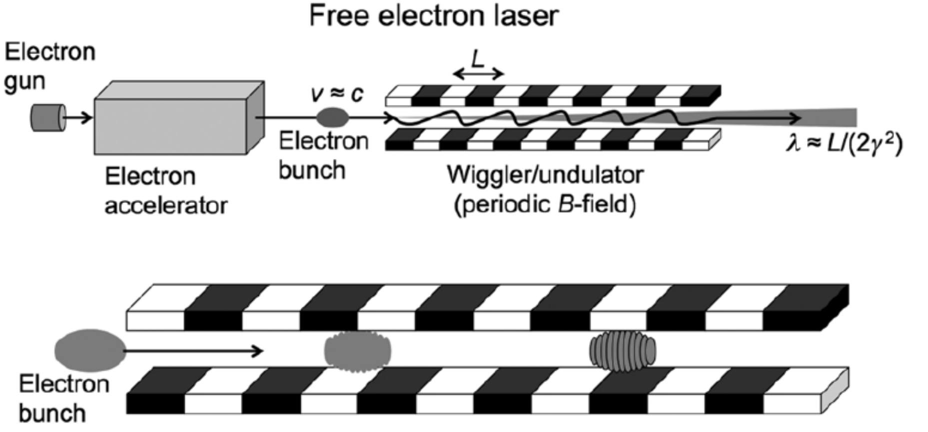
2. **Structural transitions at X-ray fluences above damage threshold**

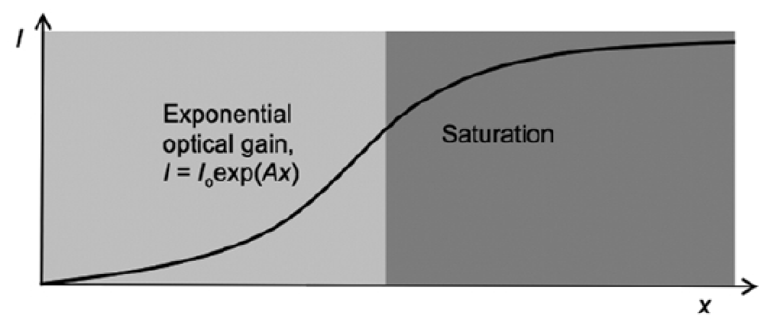
3. **X-ray induced transition to WDM state**

FELs: 4th generation light sources



photon-science.desy.de





[Ribic, Margaritondo, J. Phys. D **45** 213001 (2012)]

Pulse duration ~ down to 10 fs
Wavelength ~ VUV- hard X-ray

[This slide courtesy of Z. Jurek]

Interaction of X rays with materials:

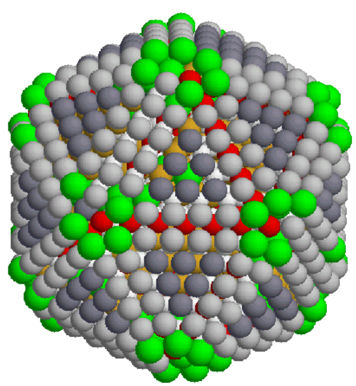
X-ray photons: elastic scattering, Compton scattering, photoionization (valence band, inner-shell), Auger & fluorescence decays

Electrons: collisional ionization and recombination from/to bands, thermalization through electron-electron interaction

Ions: electrostatic repulsion

↓ ↓ ↓

ELECTRONIC AND STRUCTURAL DAMAGE!



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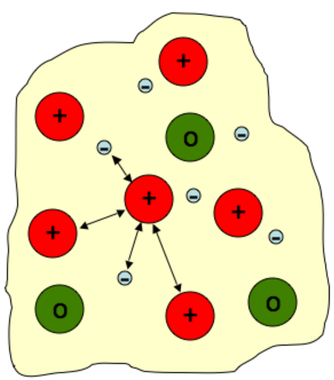


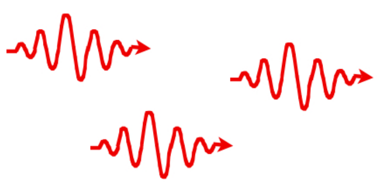
Structural transitions in solids induced by intense femtosecond XUV & X-ray pulses

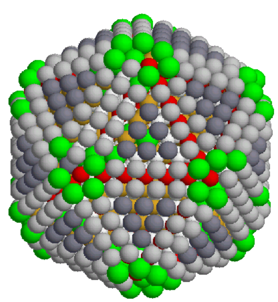
Transition depends on the average absorbed dose ....

- **<u>low dose</u> → electron excitation and relaxation → optical, magnetic changes**

- **<u>dose around structural damage threshold</u> → structural modifications**

- **<u>high dose</u> → ultrafast melting → WDM and plasma**





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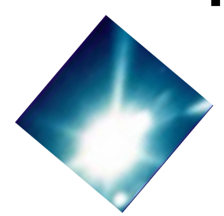

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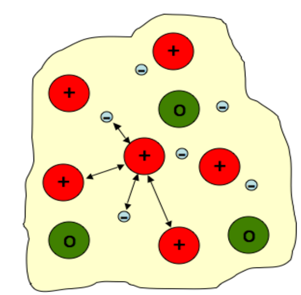

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

DESY

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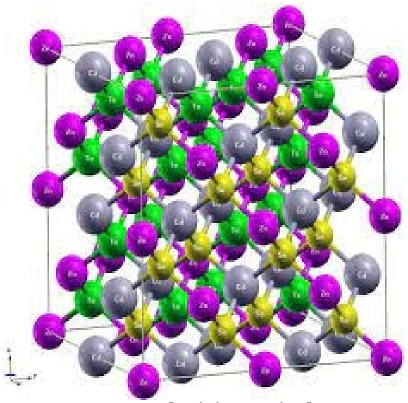
Hybrid modeling tool:
XTANT/XTANT+ code

- Dynamics of nuclei ← *Molecular dynamics*
- Excitation and relaxation of high energy free electrons and core holes ← *Monte Carlo approach*
- Low energy electrons within the valence and conduction bands ← *instantaneous thermalization assumed*
- Changes of band structure in response to nuclei dislocations ← *transferable tight binding or DFTB+ module; potential energy surface* used to derive forces
- Scattering/ionization rates *calculated using tables* or *from transient complex dielectric function*
- Electron-ion coupling ← *Boltzmann collision integral*



[Scielo Mexico]

[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]

Structural transformations induced by X-ray pulses

Damage Threshold

Non-thermal transformation (~100 fs)

↑

Change of interatomic potential induced by presence of many excited electrons

Thermal melting (~ ps)

↑

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

Melting Threshold

Simulations of both possible with our in-house hybrid code XTANT or XTANT+

[N. Medvedev, Viktor Tkachenko, V. Lipp, Zheng Li and B. Ziaja, 4Open 1,3 (2018)
V. Lipp et al., Sci. Rep. 12, 1551 (2022)]

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

CFEL SCIENCE

DESY.

Example: X-ray induced graphitization of diamond

Damage threshold

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; $sp^3 \rightarrow sp^2$

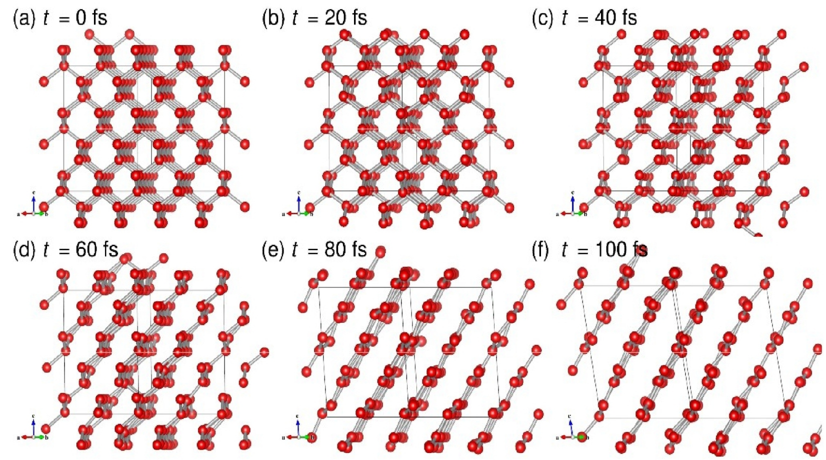
→ nuclei rebind to form an (overdense) graphite structure

→ overdense graphite relaxes slowly

Melting threshold

Results: Atomic snapshots

Photon energy 92 eV, FWHM = 10 fs



Ultrafast graphitization of diamond

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

[indico.cern.ch]

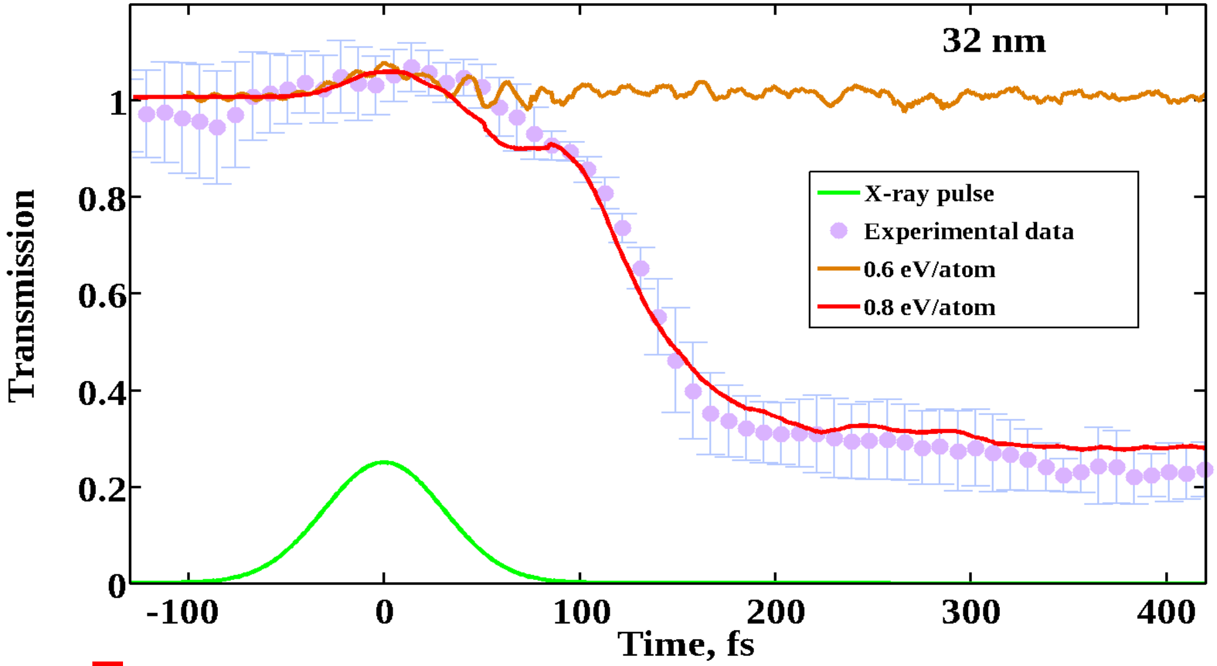
Damage thresholds ~ 1 eV/atom for various X-ray photon energies in good agreement with experiments!

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

Example: X-ray induced graphitization of diamond

Damage threshold

First indirect observation of time-resolved graphitization: transmission T(t)



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°

Melting threshold

Experiment performed by Sven Toleikis, Franz Tavella, Hauke Hoeppner, Mark Prandolini et al. at FERMI facility [F. Tavella et al., HEDP 24 (2017) 22]

-- Characteristic drop of transmission is observed during the experiment on 150 fs time-scale

Example: Atomic-scale visualization of ultrafast bond breaking in X-ray-excited diamond

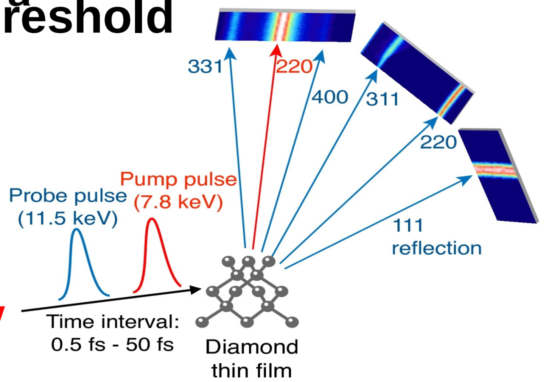
Damage threshold

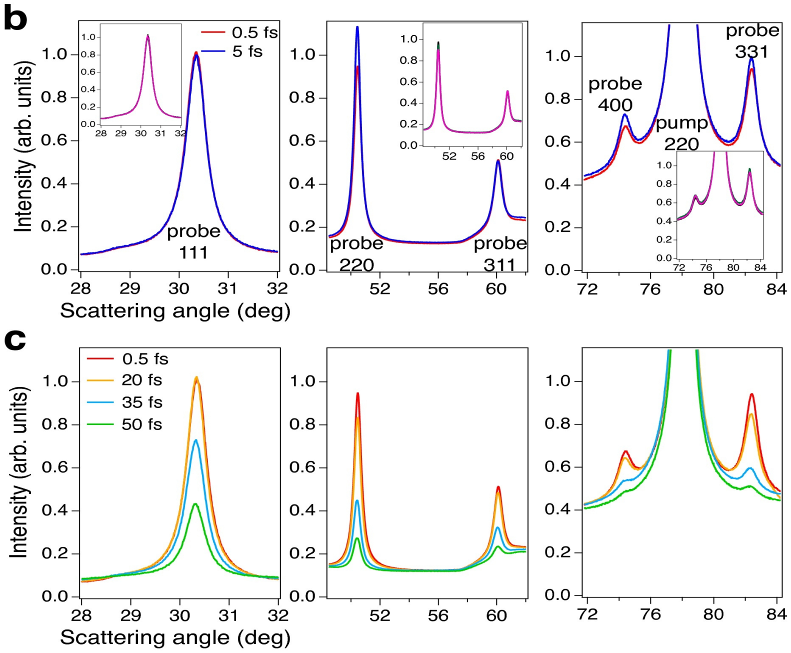
[I. Inoue et al., Phys. Rev. Lett. 126, 117403 (2021)]

X-ray pump - x-ray probe experiment at SACLA

Melting threshold

a





Theory predictions with our in-house code XTANT

Theory Team

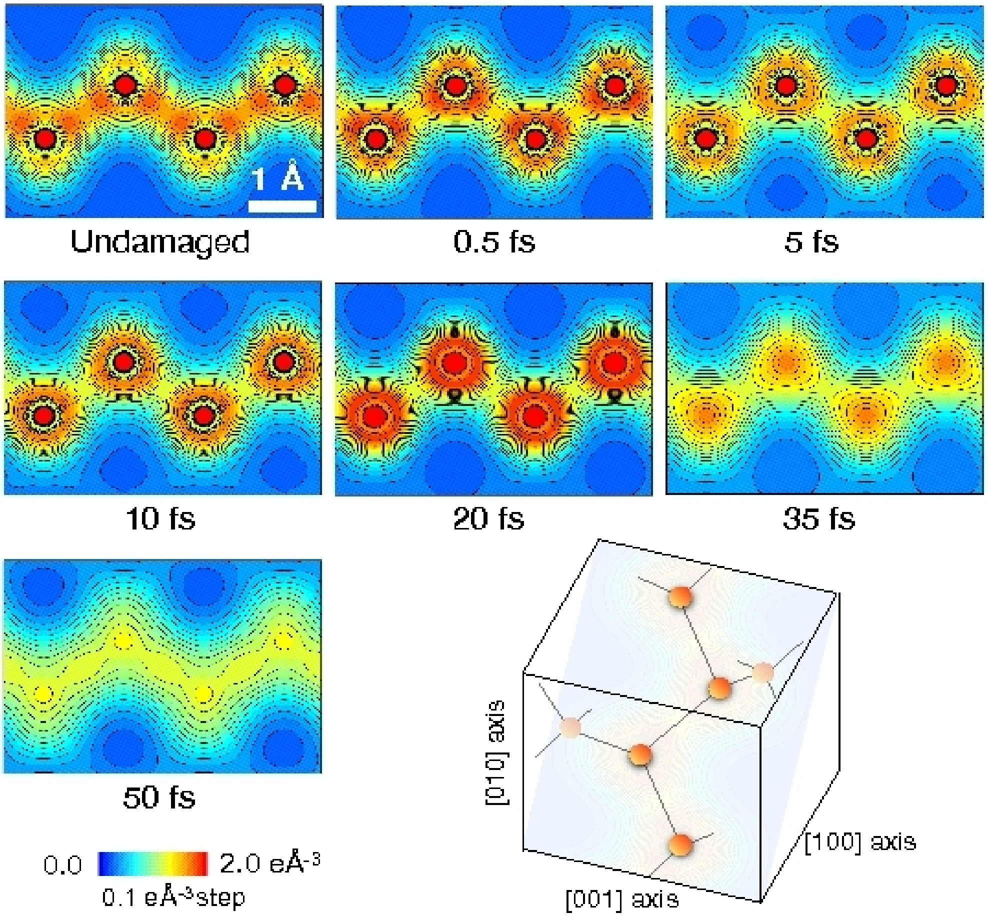
Example: Atomic-scale visualization of ultrafast bond breaking in X-ray-excited diamond

Damage threshold

Melting threshold

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

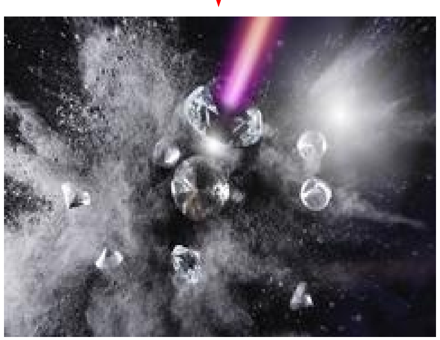
X-ray pump - x-ray probe experiment at SACLA



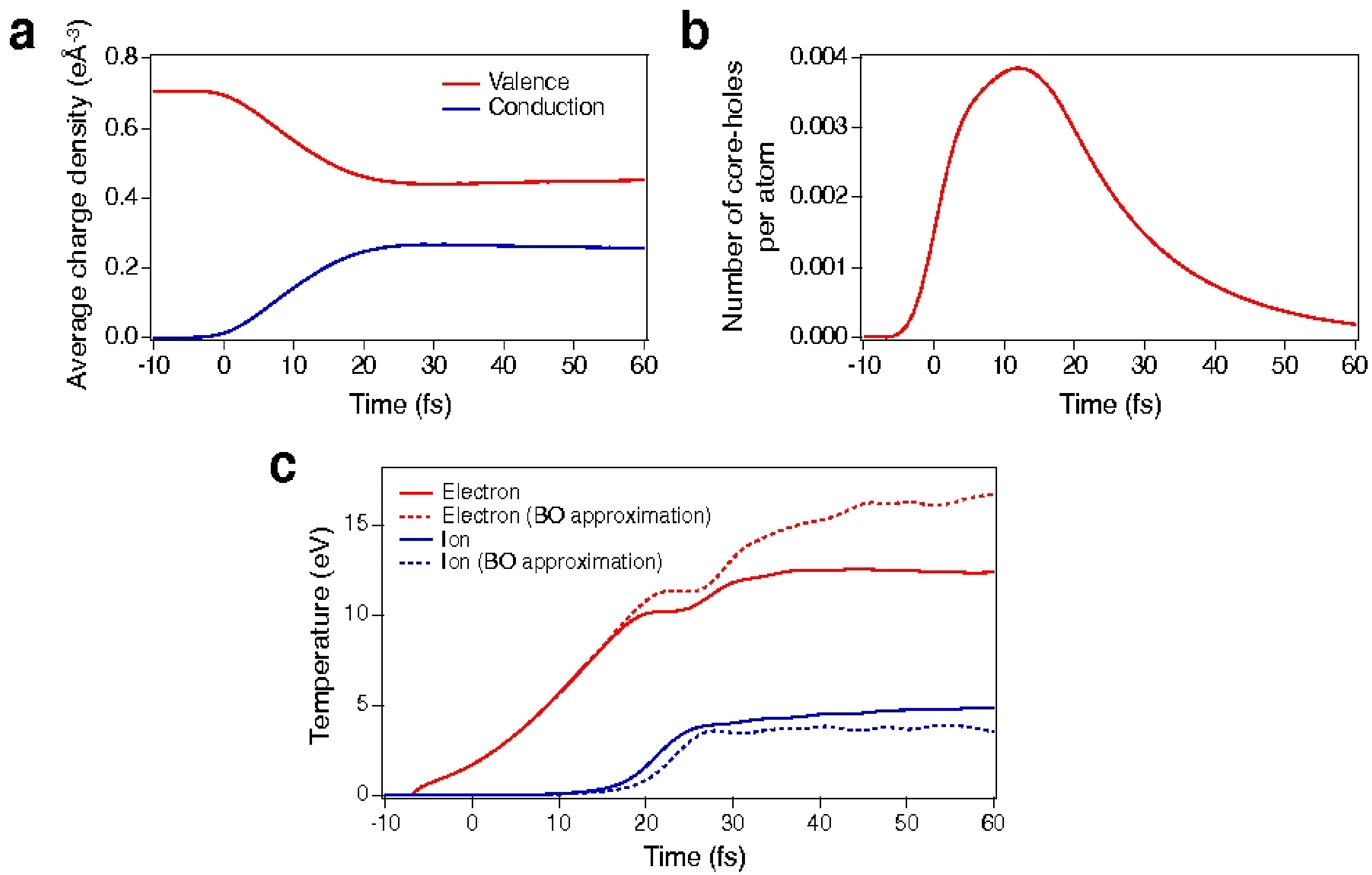
Example: Atomic-scale visualization of ultrafast bond breaking in X-ray-excited diamond

Damage threshold

Melting threshold



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



XTANT theory predictions for (a) average charge density, (b) number of K-holes & (c) electron and ion temperature → non-thermal transition

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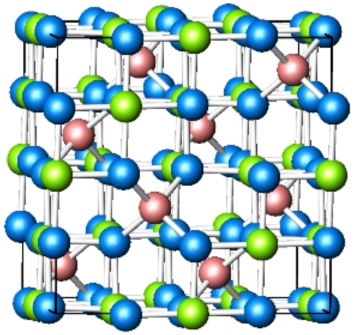
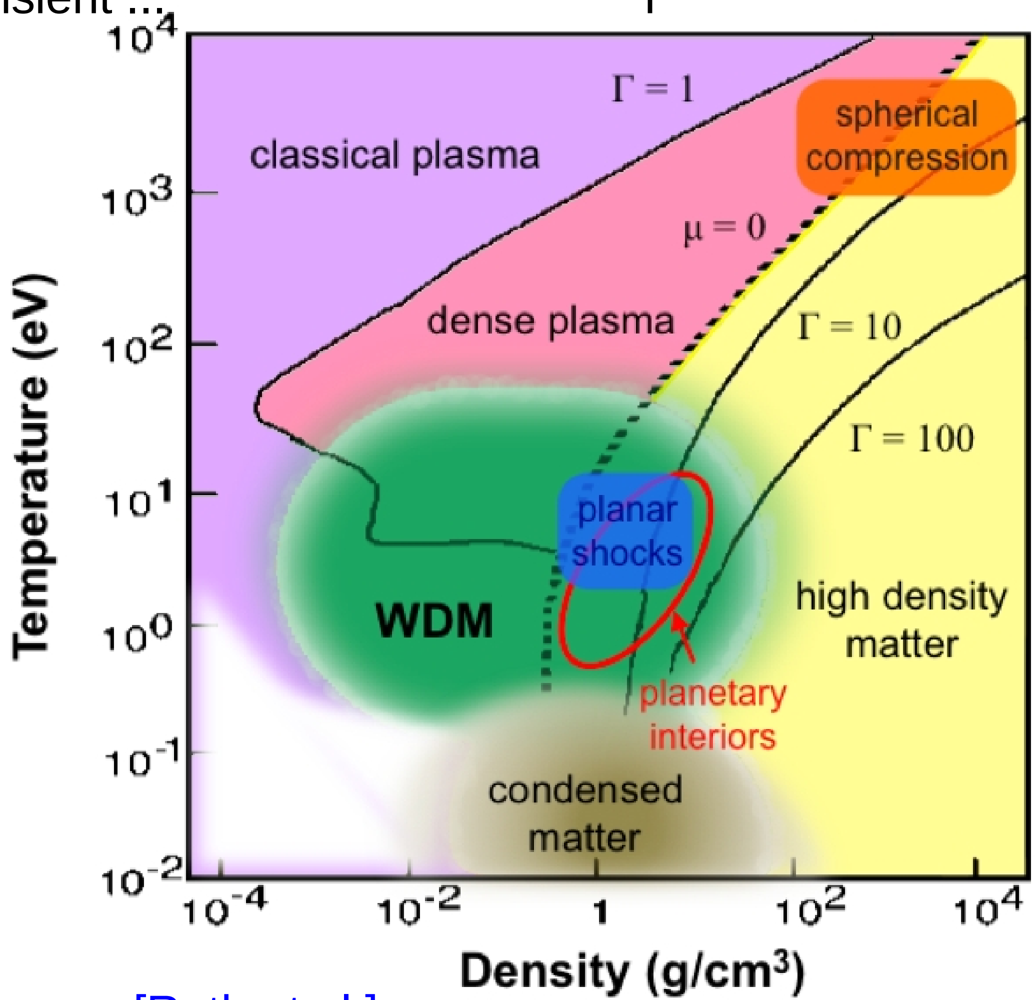
Matter in warm dense matter (WDM) state

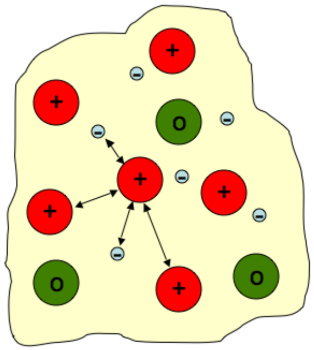
Located between solid state and plasma state. Because of its extreme temperatures and pressures, WDM tends to be drastically transient ...





[Roth et al.]

WDM defined by Γ, Y ≈ 1.



Γ – Coulomb coupling parameter =

potential energy/
kinetic energy

Y – degeneracy parameter =

Fermi energy/
kinetic energy

Example: Transient Absorption of Warm Dense Copper created by an X-Ray Free-Electron Laser

Laurent Mercadier (1,*), Andrei Benediktovitch (2), Spela Krusic (3) , Justine Schlappa (1), Marcus Agaker (4,5), Robert Carley (1), Giuseppe Fazio (6), Natalia Gerasimova (1), Young Yong Kim (1), Loic Le Guyader (1), Giuseppe Mercurio (1), Sergii Parchenko (1), Jan-Erik Rubensson (4), Svitozar Serkez (1), Michal Stransky (1,7), Martin Teichmann (1), Zhong Yin (1), Matjaz Zitnik (3), Andreas Scherz (1), **Beata Ziaja (2,7,*)**, and **Nina Rohringer (2,8)**

(1) European XFEL, 22869 Schenefeld, Germany
(2) Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany
(3) Jozef Stefan Institute, 1000 Ljubljana, Slovenia
(4) Department of Physics and Astronomy, Uppsala University, P.O. Box 516, SE-751 20 Uppsala, Sweden
(5) MAX IV Laboratory, Lund University, PO Box 118, SE-22100 Lund, Sweden
(6) Laboratorium fuer Physikalische Chemie, ETH Zurich, 8093 Zurich, Switzerland
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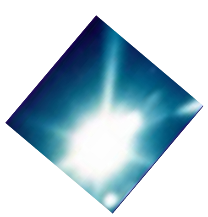
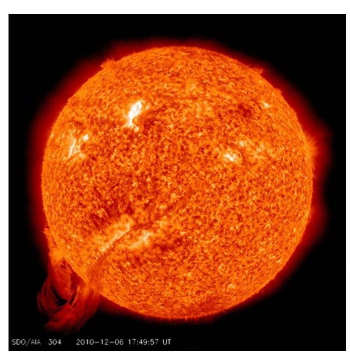
Submitted to Nat. Phys. 2023

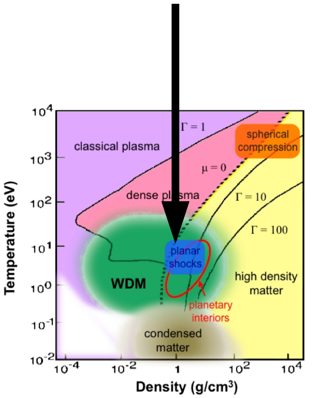




Thanking all collaborators!

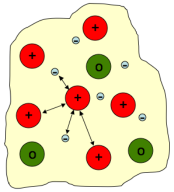
Electron kinetics in X-ray FEL excited Cu: Understanding microscopic mechanisms

'Plasma Modelling' approach



Tool:

Atomistic Boltzmann code solving classical kinetic equations for electron and ion densities. Follows non-equilibrium evolution from neutral sample to plasma and WDM state



Excitation and relaxation processes included in the kinetic equations:

- Photoionization (valence and core electrons),
- Auger decays of core holes,
- Electron-ion elastic collisions
- Collisional ionization by electrons
- Three-body recombination,
- Coulomb interactions of charges (long- and short-range)

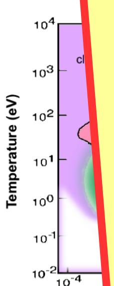
[Ziaja et al., EPJD 2006, PRL 2009, PRE 2016, EPJD 2021]

Electron kinetics in X-ray FEL excited Cu: Understanding microscopic mechanisms

Tool:

Atomistic Boltzmann code solving classical kinetic equations for electron and ion densities. Follows non-equilibrium evolution from neutral sample to plasma and WDM state

'Plasma Modelling'



Why?

Very high photon energies and electron temperatures!

Non-equilibrium electron evolution!

...onization (valence and core electrons),
- Auger decays of core holes,
- Electron-ion elastic collisions
- Collisional ionization by electrons
- Three-body recombination,
- Coulomb interactions of charges (long- and short-range)

[Ziaja et al., EPJD 2006, PRL 2009, PRE 2016, EPJD 2021]

DESY

Solving Boltzmann equations

- Classical Boltzmann kinetic equations are derived from the reduced N-particle Liouville equations. They follow the evolution of single-particle densities in phase-space.

• We adapted them to model X-ray irradiated solid or plasma systems, assuming that these samples are built of ions in various atomic configurations and of free electrons (atomistic approximation). Each of these constituents is represented by a classical phase-space density.

- The resulting set of kinetic equations for the electron distribution in phase-space, $\boldsymbol{\rho_{(e)}}$, and for distributions of various atomic configurations, $\boldsymbol{\rho_{(i,j)}}$, is:

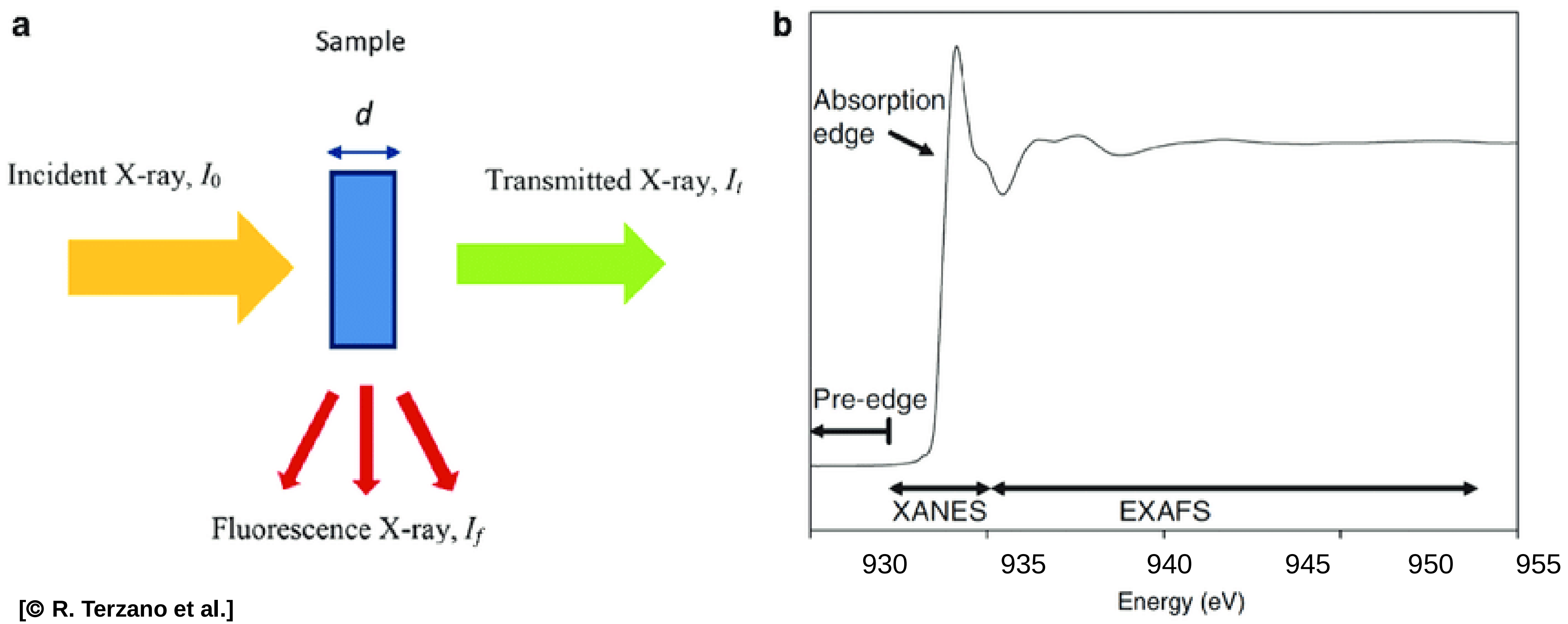
$$\partial_t \rho_{(e)}(r,v,t) + v \cdot \partial_r \rho_{(e)}(r,v,t) \ - \ F_{EM}(r,v,t)/m \cdot \partial_v \rho(e)(r,v,t) = \Omega_{(e)}(\rho_{(e)},\rho_{(i,j)},r,v,t),$$

$$\partial_t \rho_{(i,j)}(r,v,t) + v \cdot \partial_r \rho_{(i,j)}(r,v,t) + i \cdot F_{EM}(r,v,t)/M \cdot \partial_v \rho(i,j)(r,v,t) = \Omega_{(i,j)}(\rho_{(e)},\rho_{(i,j)},r,v,t).$$

The index, i=0 ,.., N_J, denotes the ion charge (with N_J being the highest charge state in the system), and the index, j=0,.., $N_C(i)$, denotes the active configuration number (with $N_C(i)$ being the maximal number of ion configurations considered for a fixed ith ion charge). Electron and ion masess are, m, and, M,
respectively.

[Ziaja et al., EPJD 2006, PRL 2009, PRE 2016, EPJD 2021]

Creating and probing warm dense matter copper with X-ray absorption at L_3-edge



[© R. Terzano et al.]

[Mercadier, BZ et al., submitted 2023, preprint: https://assets.researchsquare.com/files/rs-2396961/v1/cbedc522b1aaed26041bdde7.pdf?c=1673895193]

Material and X-ray pulse parameters:

100 nm thick Cu foil

XFEL pulse tuned to 932 eV

Beam focus ~ 4 um

Pulse duratiom ~ 15 fs FWHM

Beam energy up to 2 mJ

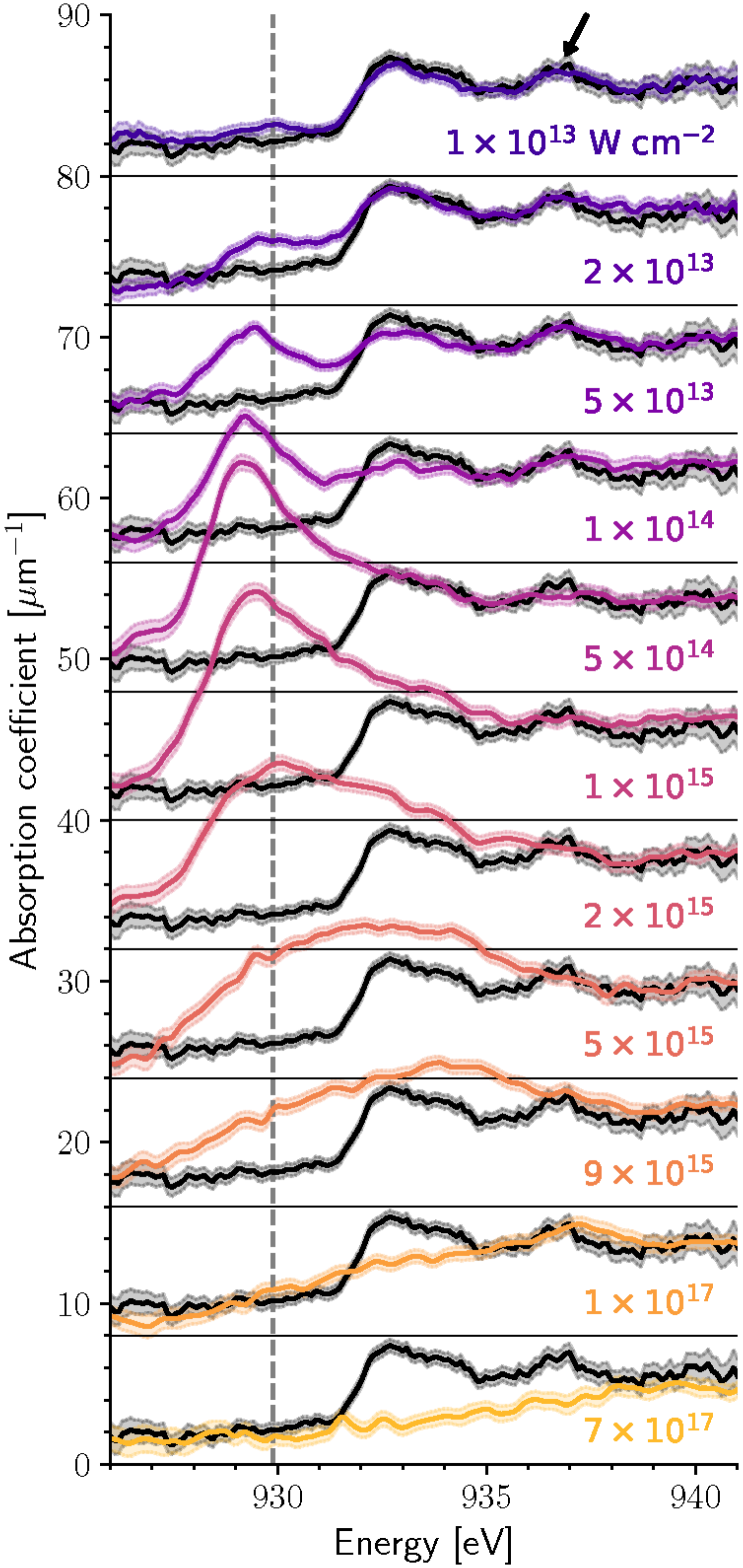
Transient XAS spectra (during the X-ray pulse):

- They are shown for various XFEL intensities. The spectra are offset from each other by 8 μm^{-1} and overlaid with a reference spectrum obtained at I = 5×10^{12} W cm−2 (black line).

- The shaded areas around each spectrum indicate the 95% confidence interval. The grey dashed line indicates the peak position of the pre-edge feature at low intensity.

- The arrow shows the characteristic fcc peak resulting from a van Hove singularity.

[Mercadier, BZ et al., submitted 2023, preprint: https://assets.researchsquare.com/files/rs-2396961/v1/cbedc522b1aaed26041bdde7.pdf?c=1673895193]



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European XFEL

Experiment

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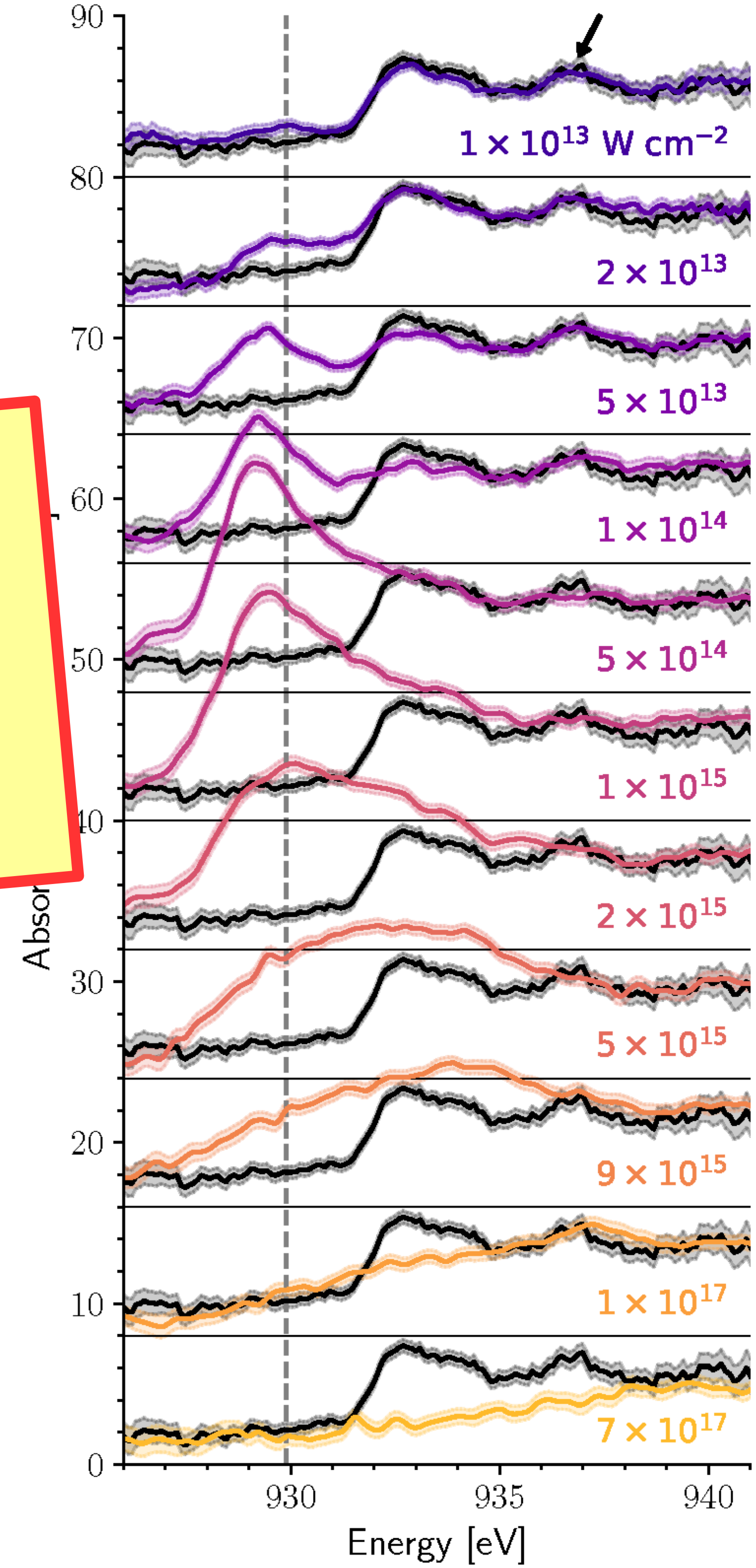
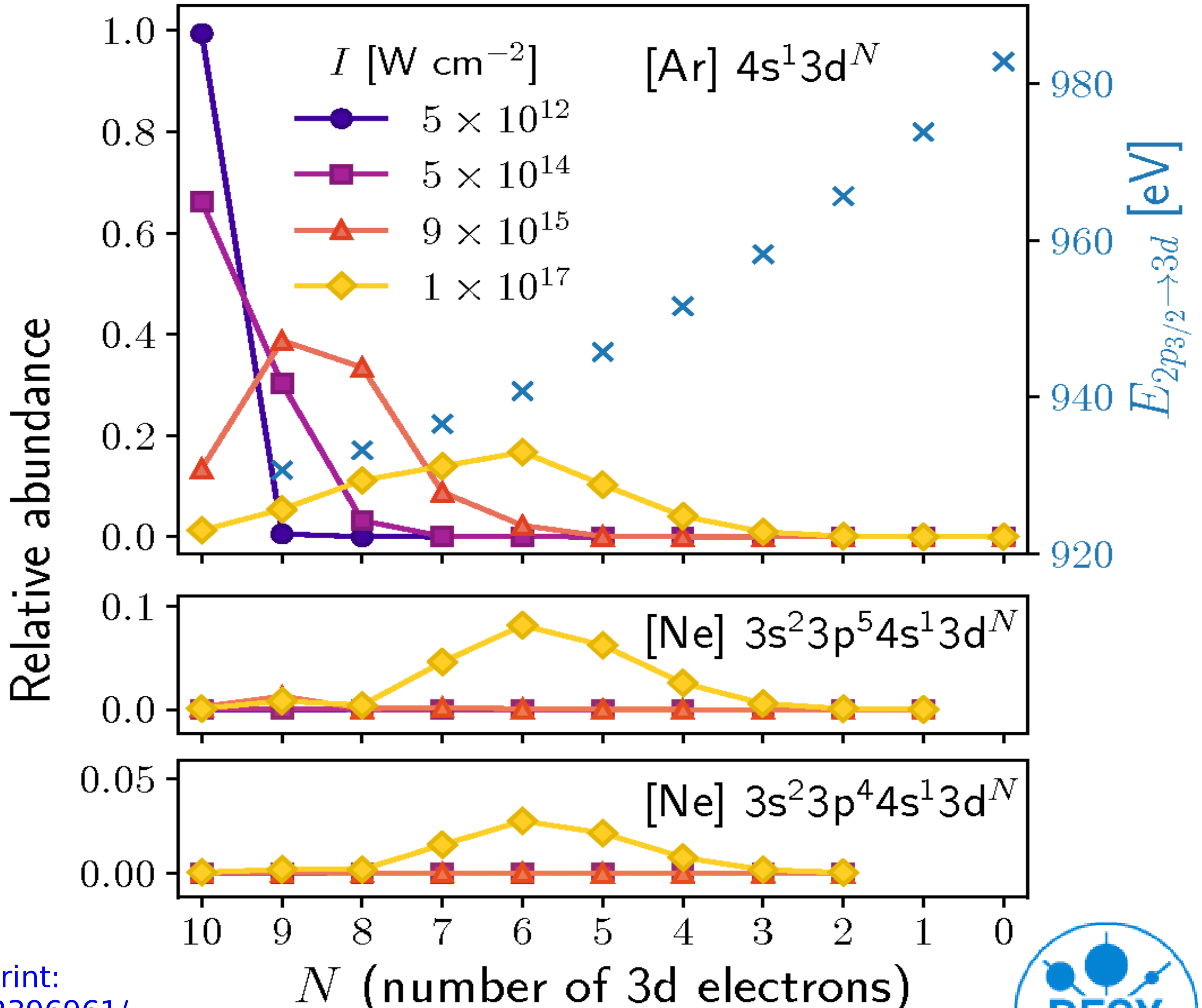


Creating and probing warm dense matter copper with X-ray absorption at L_3-edge

Relative abundance of the most populated copper configurations for different pulse intensities calculated with the Boltzmann kinetic model.

The base configuration is shown on each panel and the number of 3d electrons N is varied.

The blue crosses and right axis in the upper panel show the calculated energy of the $2p_{3/2} \rightarrow 3d$ transition as a function of N.



[Mercadier, BZ et al., submitted 2023, preprint: https://assets.researchsquare.com/files/rs-2396961/v1/cbedc522b1aaed26041bdde7.pdf?c=1673895193]

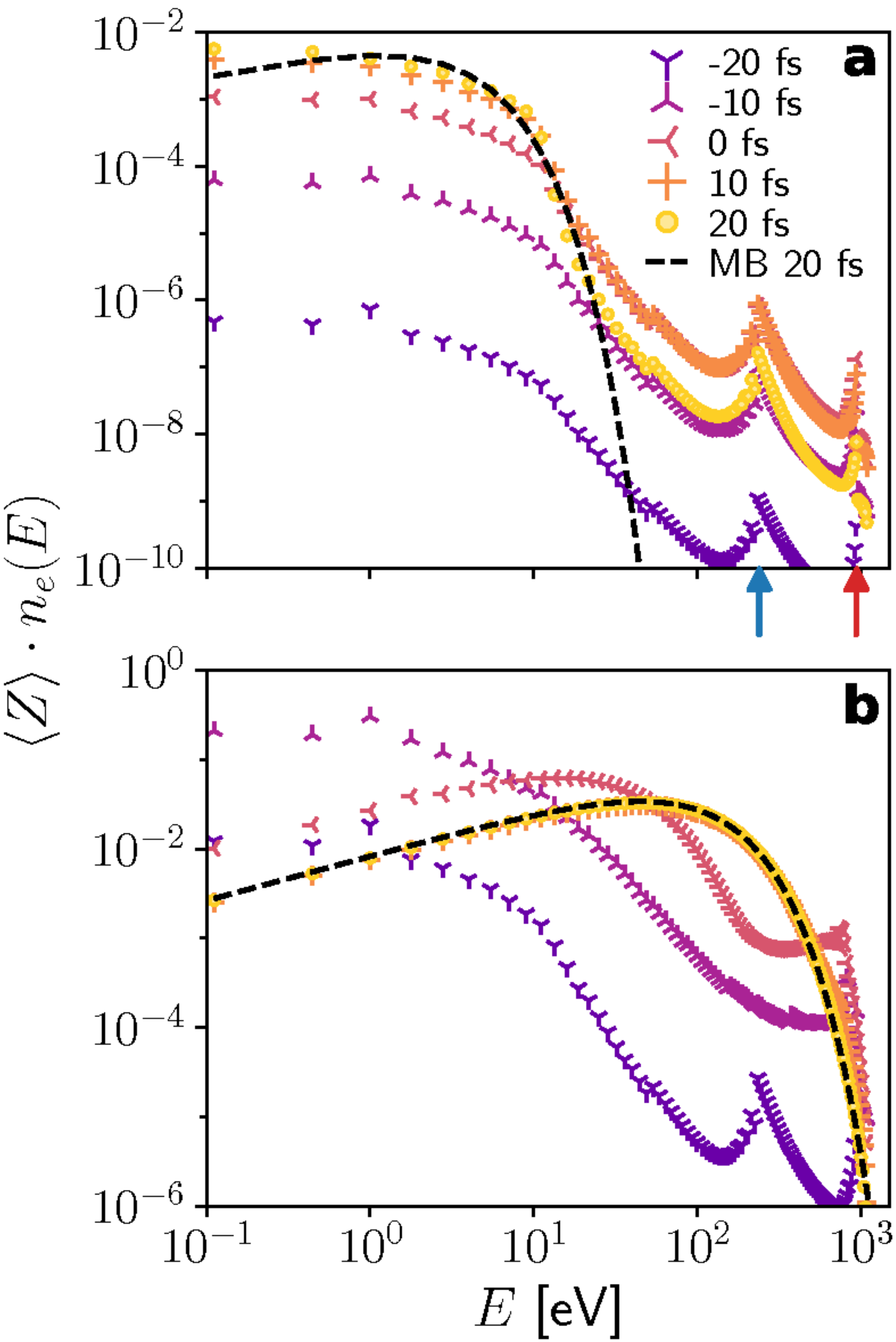
Rich electron dynamics following the X-ray irradiation with:

(a) pulse intensity I = 5 × 10^{12} W cm^{-2},
(b) pulse intensity I = 1 × 10^{17} W cm^{-2}.

- The dashed curves show the Maxwell-Boltzmann distributions with a kinetic temperature equal to the average electron kinetic energy at 20 fs.

- The broad peak with substructure at around 250 eV indicated by a blue arrow corresponds to the secondary electron emission during the collisional ionization of neutral Cu and Cu+ ground states.

- The sharp peak at 943 eV indicated by a red arrow is due to the Auger decay of the $2p_{3/2}$ core hole.



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Rich electron dynamics following the X-ray irradiation with:

(a) pulse intensity I = 5 × 10^{12} W...
(b) pulse intens...

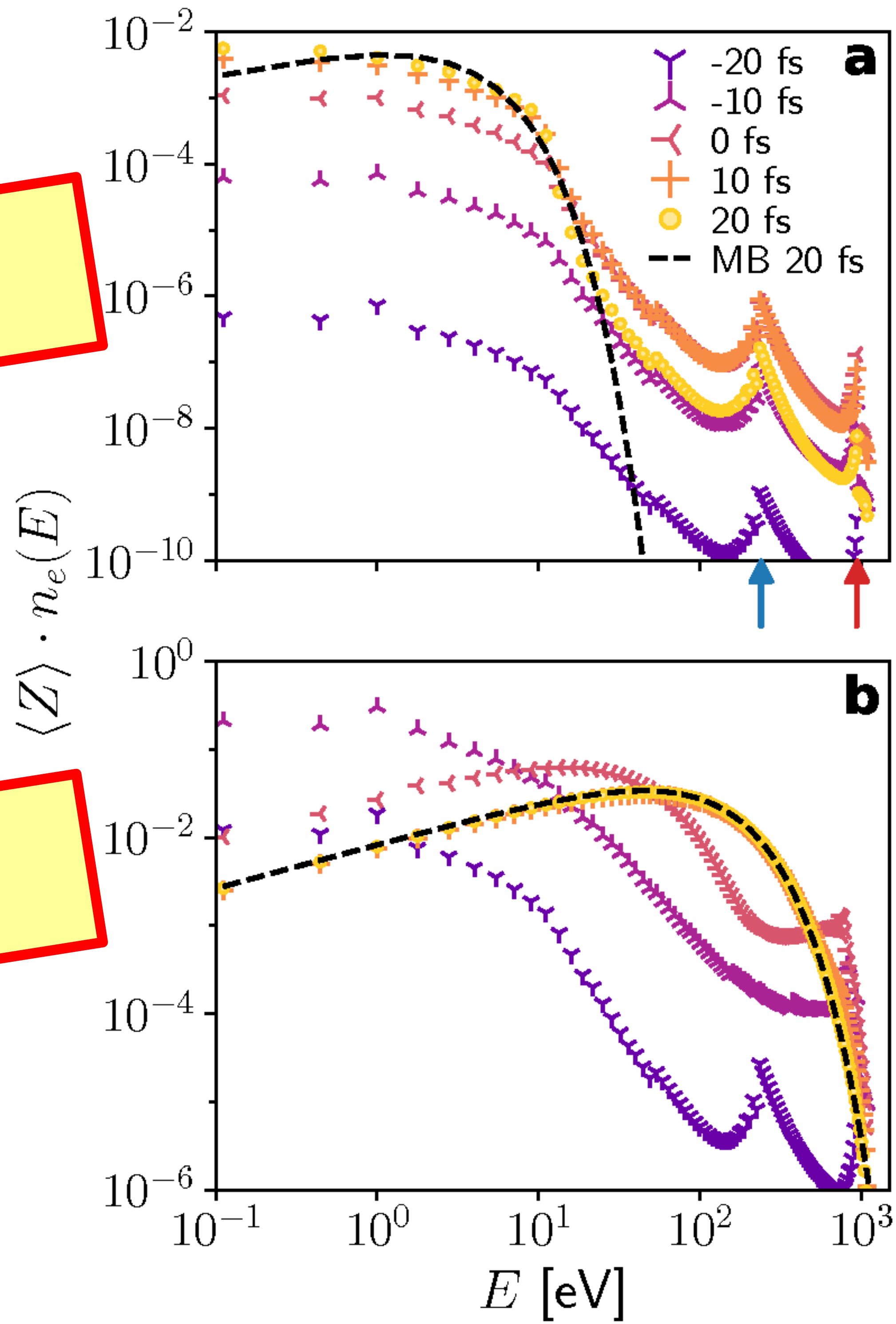
Non-thermal electron distribution

- The dashed curve ... Maxwell-Boltzmann distributions with a kinetic temperature equal to the average electron kinetic energy at 20 fs.

- The broad peak with substructure at around 250 eV indicated by a blue arrow corresponds to the secondary electron emission during the collisional ionization of neutral Cu and Cu+ ground states.

- The sharp peak a... arrow is due to the ... hole.

Thermal electron distribution



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Creating and probing warm dense matter copper with X-ray absorption at L_3-edge

Conclusions:

- Transient XAS reveals the richness of the non-equilibrium electron dynamics within the material on the timescale of the X-ray pulse duration (here 15 fs FWHM).

- At moderate intensities, the localized character of the excited Cu 3d band and its negative energy shift is observed

- At higher intensities, the transition to saturable absorption regime occurs, through which metal becomes transiently "transparent" in specific spectral regions → effective "shortening" of pulse duration

- Agreement between the data and the Boltzmann model reasonable. Standard DFT models not applicable here → possible benchmark for future DFT developments

- Time resolution of the XAS method is only limited by the pulse duration. Attosecond XFEL pulses will widen the applicability of the presented approach for studies of matter under extreme conditions.

[Mercadier, BZ et al., submitted 2023, preprint: https://assets.researchsquare.com/files/rs-2396961/v1/cbedc522b1aaed26041bdde7.pdf?c=1673895193]

Summary

(1) Transitions in materials induced by X-ray radiation:

-below structural damage threshold – non-equilibrium electron kinetics

-below melting threshold – also rearrangement of atomic structure:

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

(2) Diagnostics:

- surface damage

- transient optical properties

- X-ray diffraction

- X-ray absorption

CFEL-XM computational tools to describe X-ray induced transitions in materials

- **XTANT and 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 → **available under DESY license**
- **XSPIN**: a hybrid simulation tool to study X-ray induced magnetic transitions in solids
- **Boltzmann equation solver**: solves classical kinetic equations for X-ray irradiated finite-size and bulk systems, using atomistic approximation
- **SURFwiX**: models surface damage by X rays at realistic experimental conditions (micrometer +nanoseconds)
- **NanoDiff:** models electron and heat diffusion after X-ray pulse impact on um and ns timescales

https://xm.cfel.de/






K. Kapcia V. Lipp N. Medvedev (< 2016) M. Stransky V. Tkachenko B. Ziaja

CFEL-XM computational tools to describe X-ray induced transitions in materials

- **XTANT and 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 → **available under DESY license**
- **XSPIN**: a hybrid simulation tool to study X… transitions in solids
- **Boltzmann eq…** …ations for X-ray irradiated finite-size and b… …istic approximation
- **SURFwiX**: models surface damage by X rays at realistic experimental conditions (micrometer +nanoseconds)
- **NanoDiff:** models electron and heat diffusion after X-ray pulse impact on um and ns timescales

Further Code Transfer under DESY License planned.

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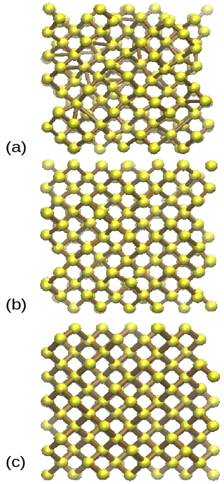
Thank you for your attention!

CFEL-XM Group

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

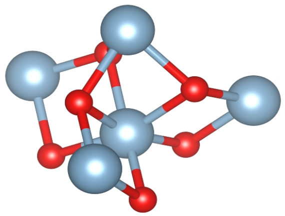
Recent examples of XTANT/+ applications



▪ Limitations of Structural Insight into Ultrafast Melting of Solid Materials with X-ray Diffraction Imaging
[V. Tkachenko et al., Appl. Sci. 11,5157 (2021)]

→ volume integration necessary to interpret imaging data from irradiated silicon crystal

▪ Observation of atomic displacements on subatomic length scales in Al_2O_3
[I. Inoue et al., PRL 128, 223203 (2022)]



→ no displacement observed up to 20 fs since time zero. High-accuracy reconstruction possible

▪ Modeling of ultrafast magnetic processes triggered by high-intensity X-ray pulses
[K. Kapcia, V. Tkachenko, B. Ziaja et al., npj Comput. Mat. 8, 212 (2022)]

→ **nanoscopic model of X-ray induced demagnetization**