Ultrafast transformation in matter induced by intense X-ray radiation - part I

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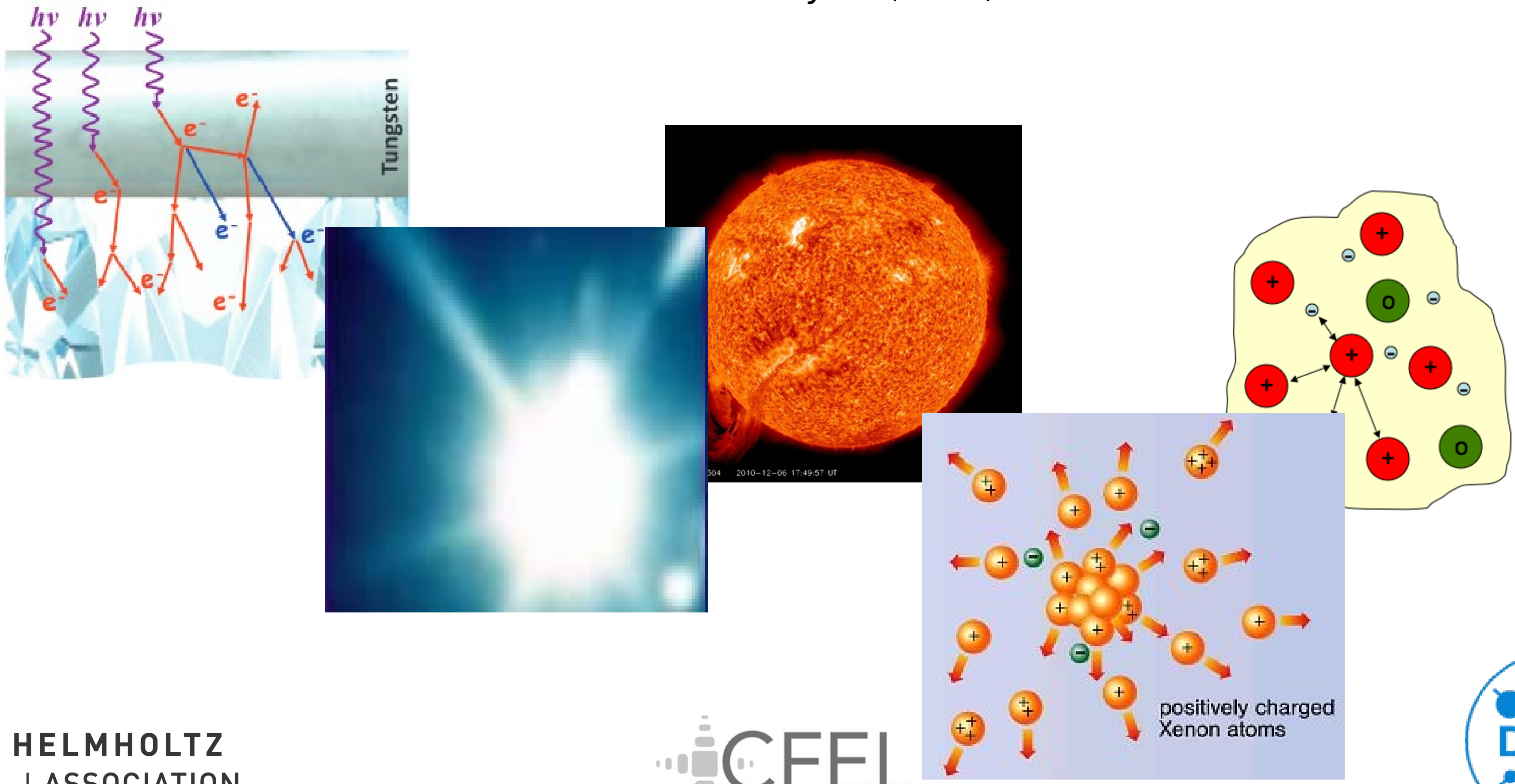

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CFEL-DESY Theory Division
at Center for Free-Electron Laser Science

The CFEL Theory Division develops theoretical and computational tools to predict the behavior of matter exposed to intense electromagnetic radiation. We employ quantum-mechanical and classical techniques to study ultrafast processes that take place on time scales ranging from 10^{-12} s to 10^{-18} s. Our research interests include the dynamics of excited many-electron systems; the motion of atoms during chemical reactions; and x-ray radiation damage in matter.

Members of the CFEL-DESY Theory Division: C. Arnold, S. Bazzi, Y.-J. Chen, O. Geffert, D. Gorelova, L. Inhester, Z. Jurek, K. Hanasaki, A. Hanna, A. Karamatskou, M. Krishna, Z. Li, M. A. Malik, N. Medvedev, P. K. Mishra, R. Santra (Division Director), V. Saxena, J. M. Slowik, S.-K. Son, V. Tkachenko, K. Toyota, O. Vendrell, B. Ziaja

3 subgroups: 'Ab-initio X-ray Physics' (R. Santra), 'Chemical Dynamics' (O. Vendrell), 'Modeling of Complex Systems' (B. Ziaja)

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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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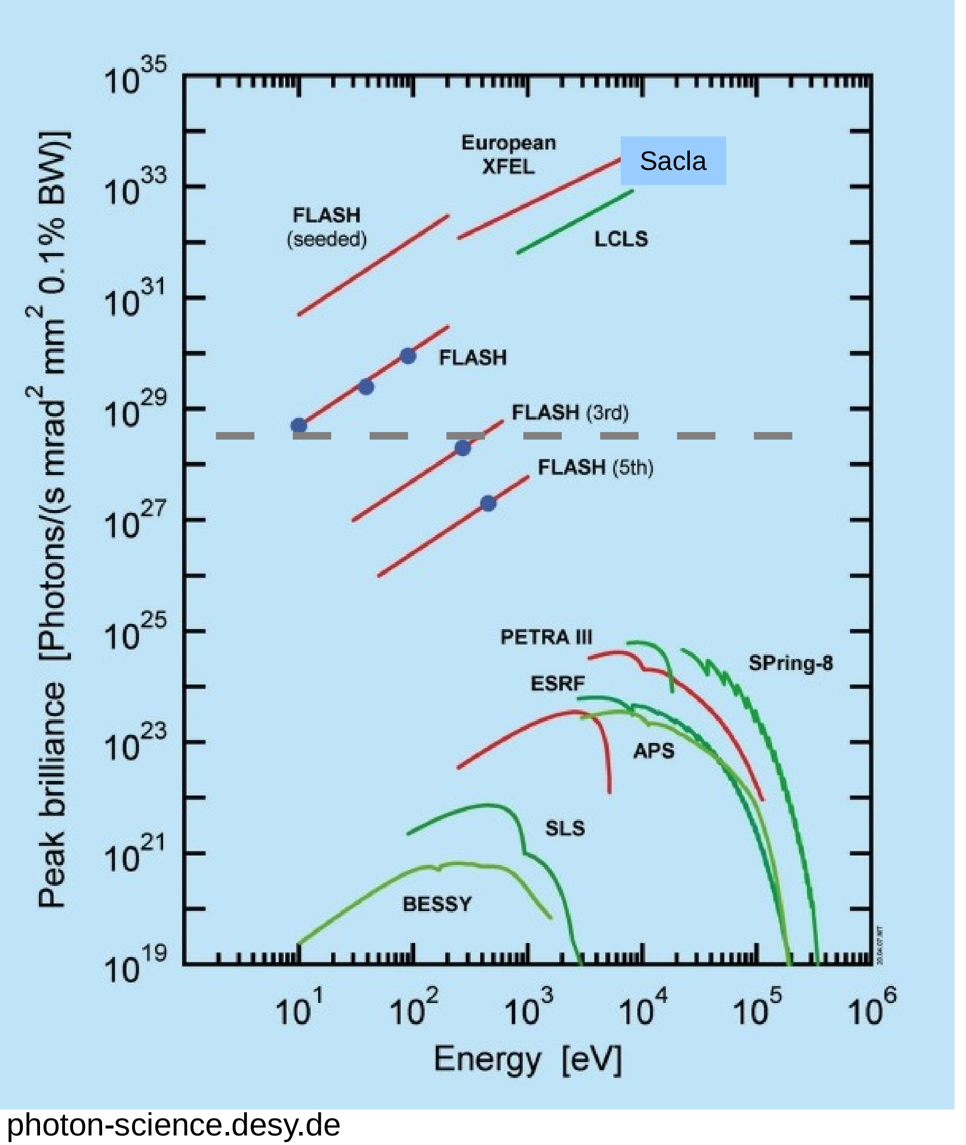
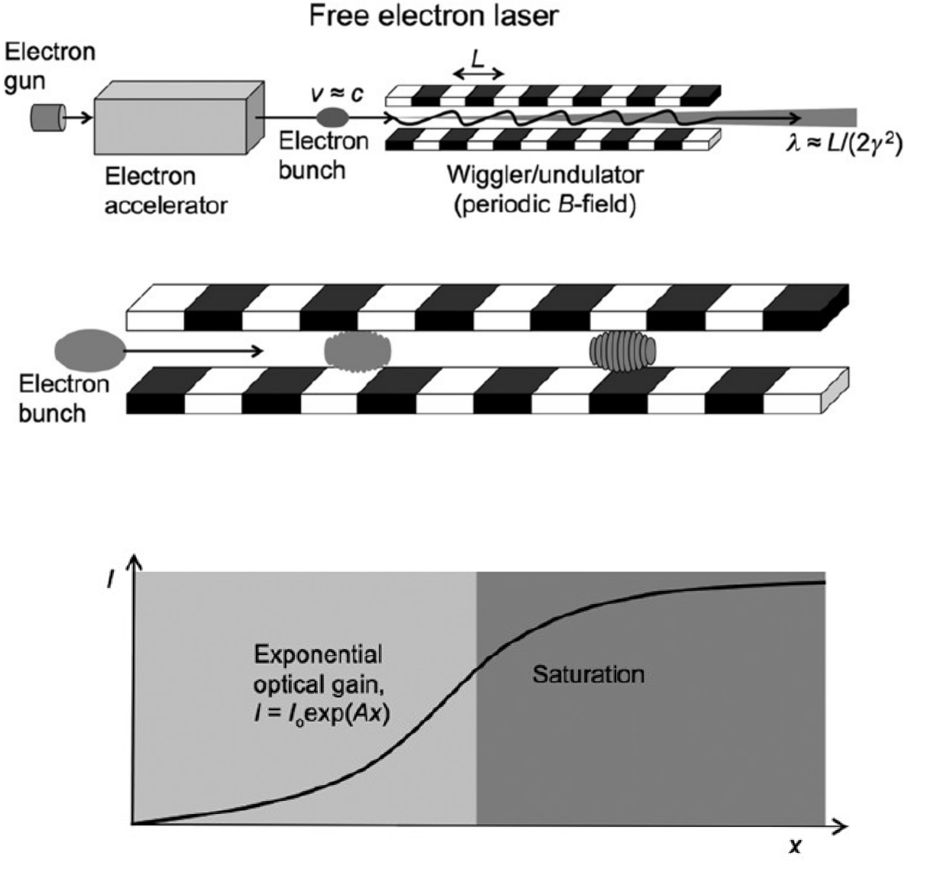
Structural transitions in solids induced by X-ray radiation

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

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]

Structural transitions in solids induced by X-ray radiation

Transition in irradiated solid depends on material properties and pulse parameters (photon energy, fluence, temporal/spatial pulse profile etc).

↓ ↓ ↓

Pulse gets absorbed. This then 'translates' into the average dose absorbed per atom.

For X-rays there is no inverse bremsstrahlung → 'easy' control of energy absorption.

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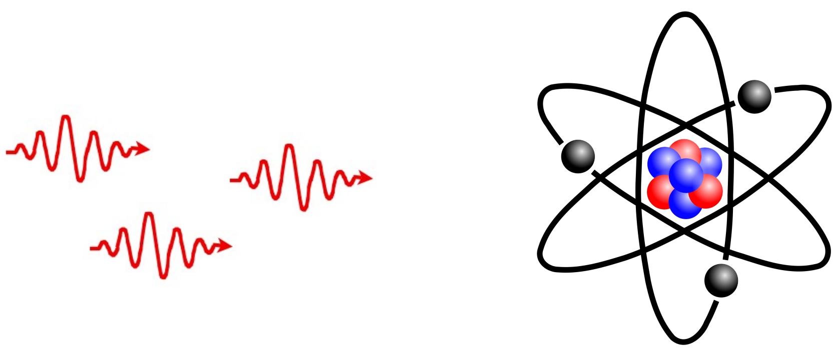
Structural transitions in solids induced by X-ray radiation

Transition depends on the average absorbed dose ....



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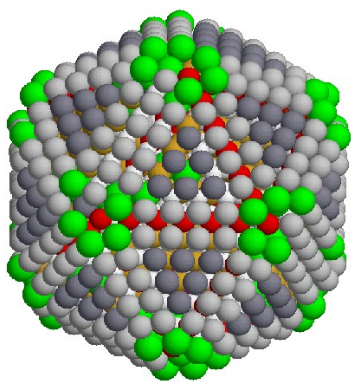
Main interactions:

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

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

Ions: electrostatic repulsion → band modification → structural transition?



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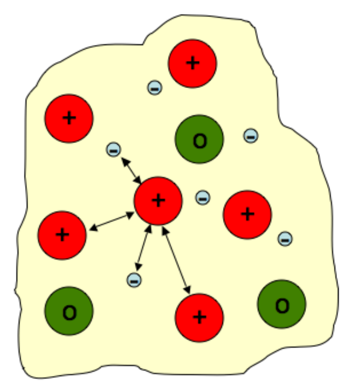
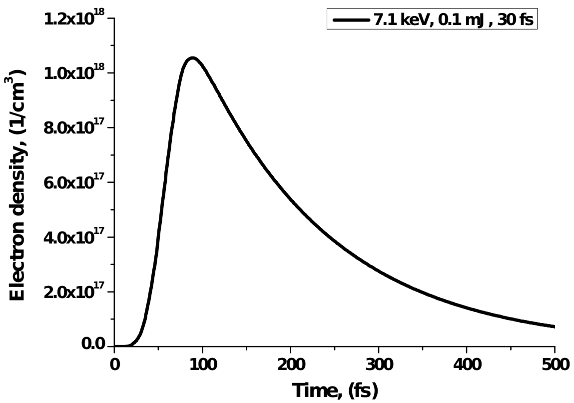


Interaction of solids with low-fluence femtosecond X-ray pulses

Low dose

Radiation excites free electrons within solids which induce transient change of solid's optical properties (reflectivity, transmission) but no structural changes.

Example:
SiO_2



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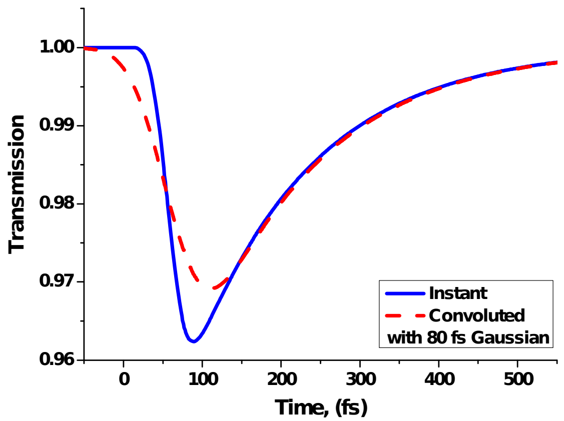

Electron density translates into transient change of optical properties with Drude model or ab-initio calculated dielectric function

[Medvedev et al., CPP 53 (2013) 347]

→ application for a non-destructive high-resolution FEL pulse timing tool

Damage Threshold

Harmand et al. (Medvedev, Ziaja), Nat. Phot. 7 (2013) 215;
Riedel et al.(Medvedev,Ziaja), Nat. Commun. 4 (2013) 1731

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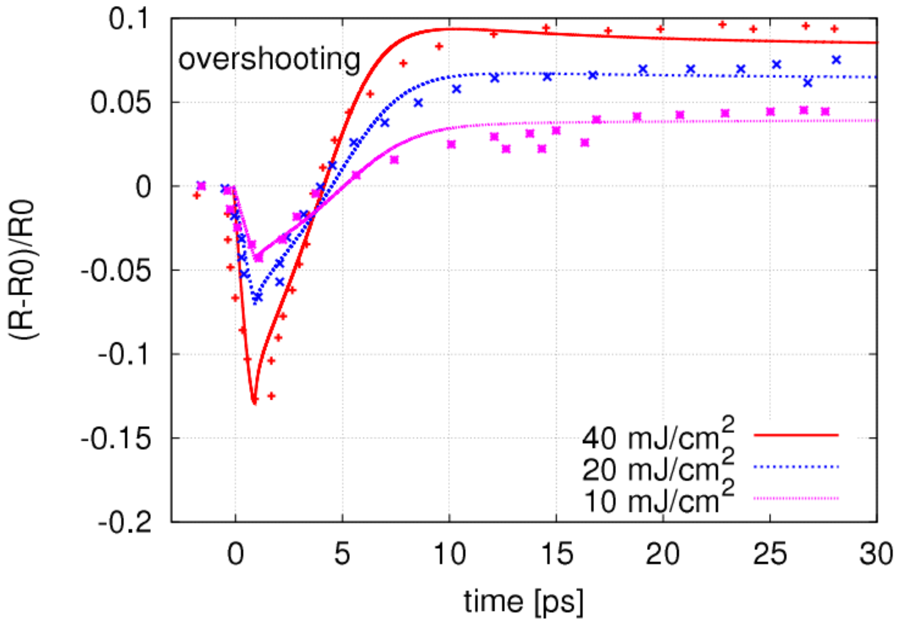
[Images courtesy of N. Medvedev]

Interaction of solids with low-fluence femtosecond X-ray pulses

Low dose

Reflectivity overshooting in GaAs



LCLS measurement (800 eV)

Damage Threshold

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[Images courtesy of V. Tkachenko]

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[B.Z., N. Medvedev, V. Tkachenko, T. Maltezopoulos, W. Wurth, *Sci. Rep*. **5**, 18068 (2015)]

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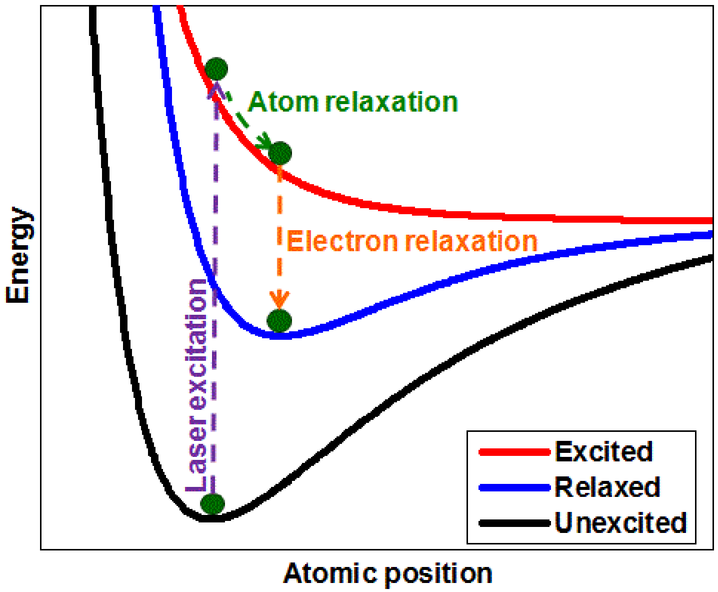
Interaction of solids with femtosecond X-ray pulses of fluence above the damage threshold

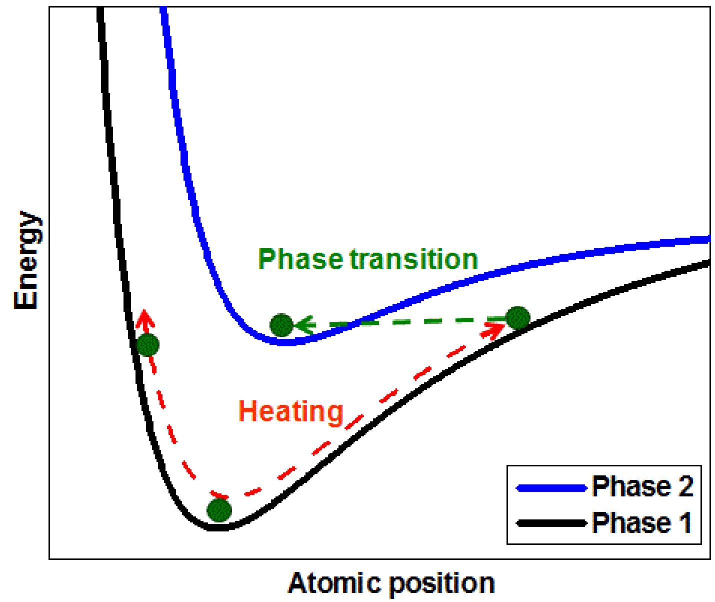
Damage threshold

Non-thermal melting (~100 fs)



Change of interatomic potential

Thermal melting (~ ps)



Heating of atomic lattice due to el-ph coupling within the same potential

Melting threshold

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

[Courtesy of N. Medvedev]

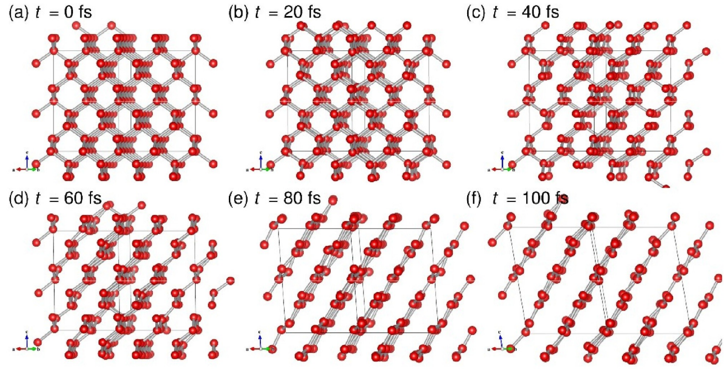
Interaction of solids with femtosecond X-ray pulses of fluence above the damage threshold

Damage threshold

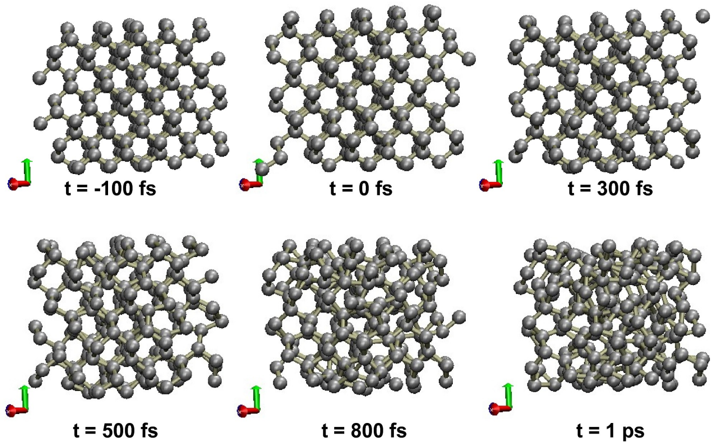
Structural transitions in solids:

→ graphitization of diamond
ultrafast non-thermal process modeled within Born-Oppenheimer scheme

Photon energy 92 eV, FWHM = 10 fs



→ amorphization of silicon
contribution of non-thermal and thermal melting (due to electron-phonon coupling); extended Born-Oppenheimer scheme



Melting threshold

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

Damage thresholds in good agreement with experiments!

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[Images courtesy of N. Medvedev]

Interaction of solids with femtosecond X-ray pulses of fluence above the damage threshold

Damage threshold

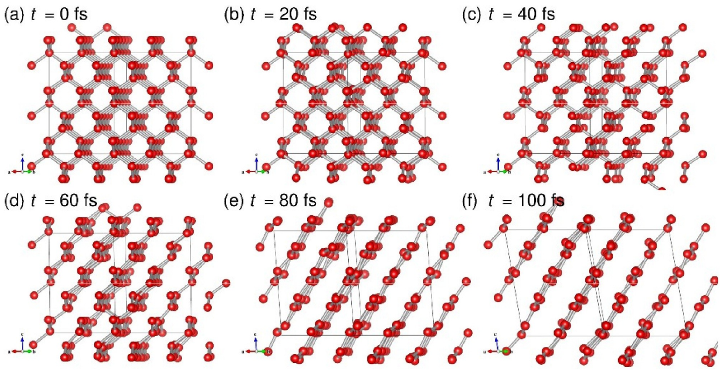
Simulations with dedicated code XTANT: X-ray induced Thermal and Non-Thermal Transitions [Medvedev et al.] →

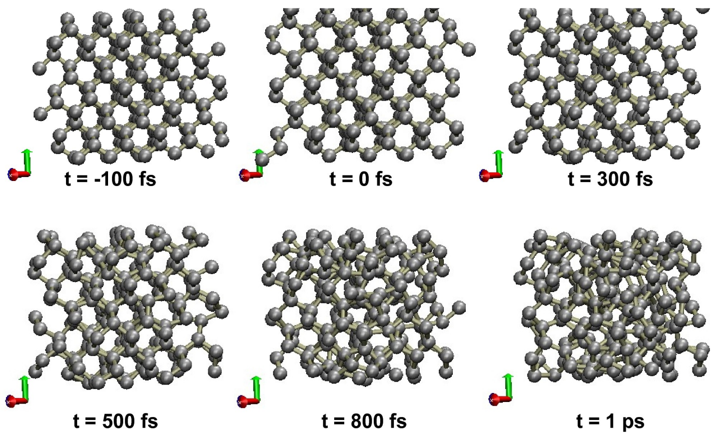
→

Melting threshold

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

Photon energy 92 eV, FWHM = 10 fs





Damage thresholds in good agreement with experiments!

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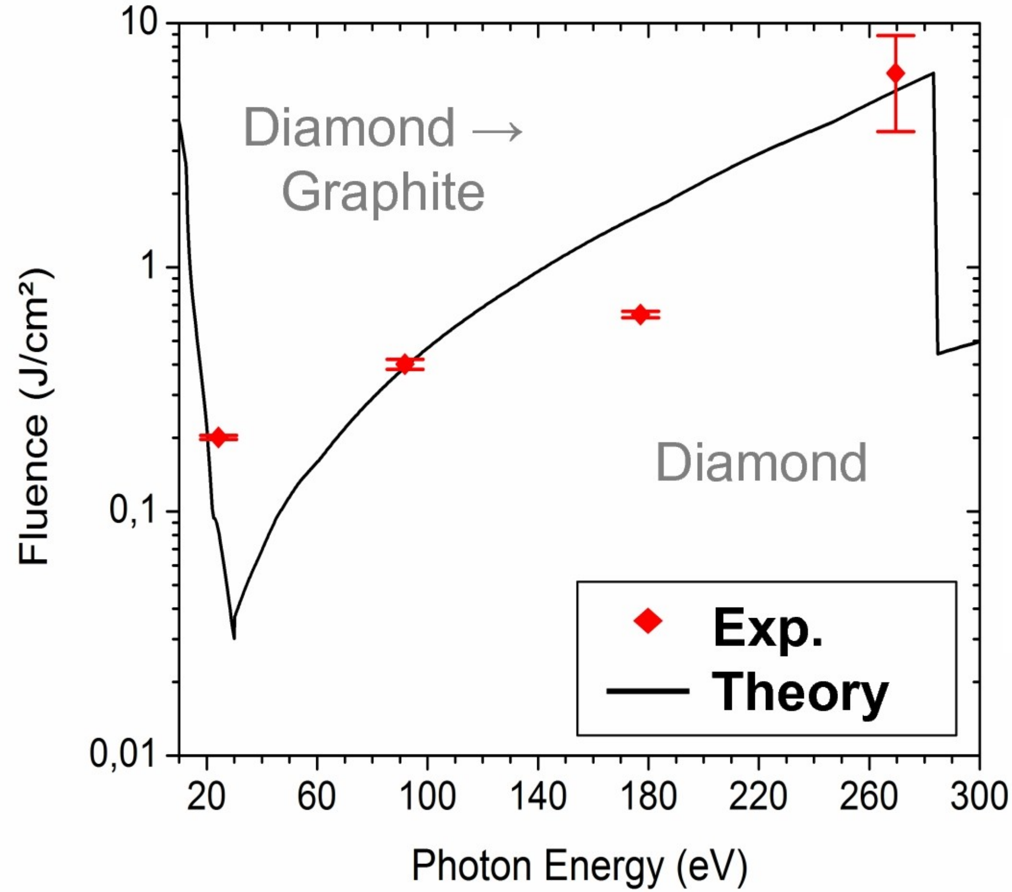
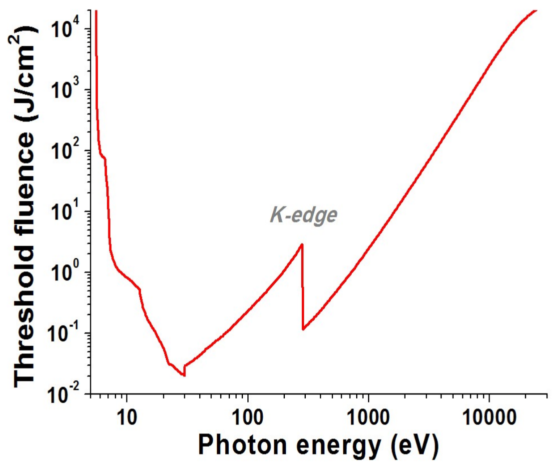
[Images courtesy of N. Medvedev]

Graphitization Damage threshold

Irradiated diamond turns into graphite if the fluence is high:





Extrapolation to hard X-rays

Damage threshold is in a good agreement with the experiments by J. Gaudin *et al.* (FLASH)

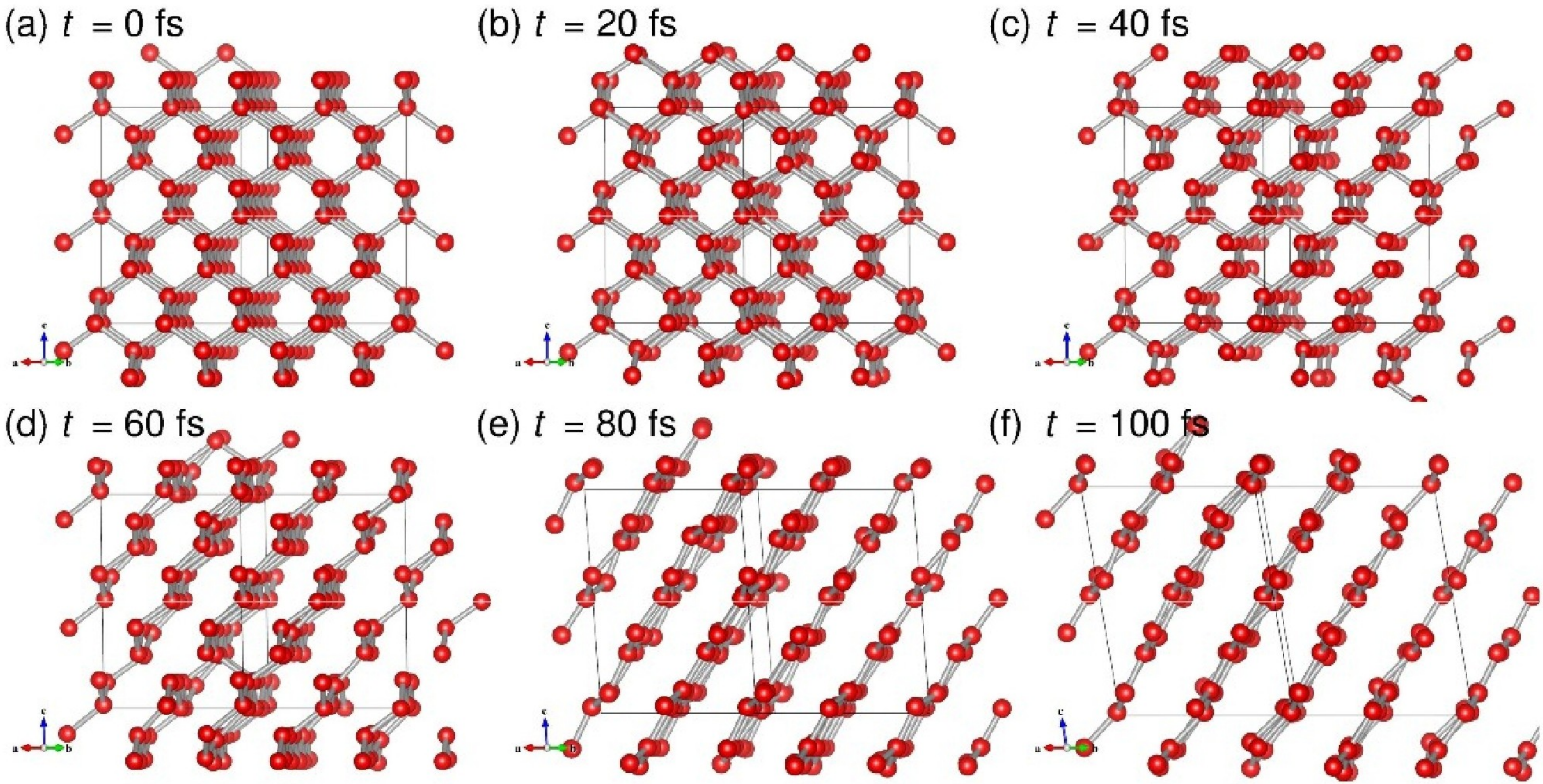
[J. Gaudin *et al.*, (2013) PRB, Rapid Comm. 88 (2013) 060101 (R)]

[N. Medvedev , H. Jeschke, BZ, PRB 88 (2013) 224304]

[This slide courtesy of

Graphitization: Atomic snapshots

Photon energy 92 eV, FWHM = 10 fs



Ultrafast graphitization of diamond

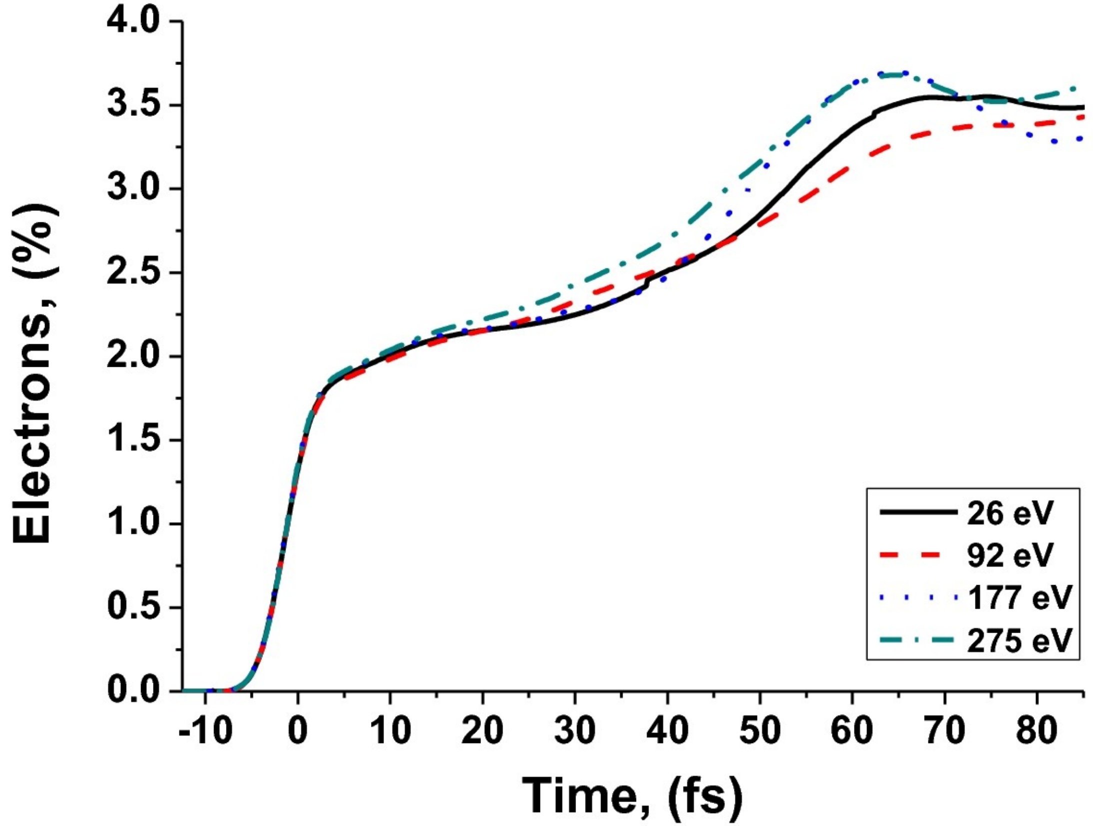
[N. Medvedev, H. Jeschke, B. Ziaja, NJP 15 (2013) 015016]

Increase of electronic density → band gap collapse

[This slide courtesy of

Results: Conduction band electrons

Different photon energies: 26 eV, 92 eV, 177 eV, 275 eV



When electron density overcomes threshold value of 1.5 %, phase transition occurs

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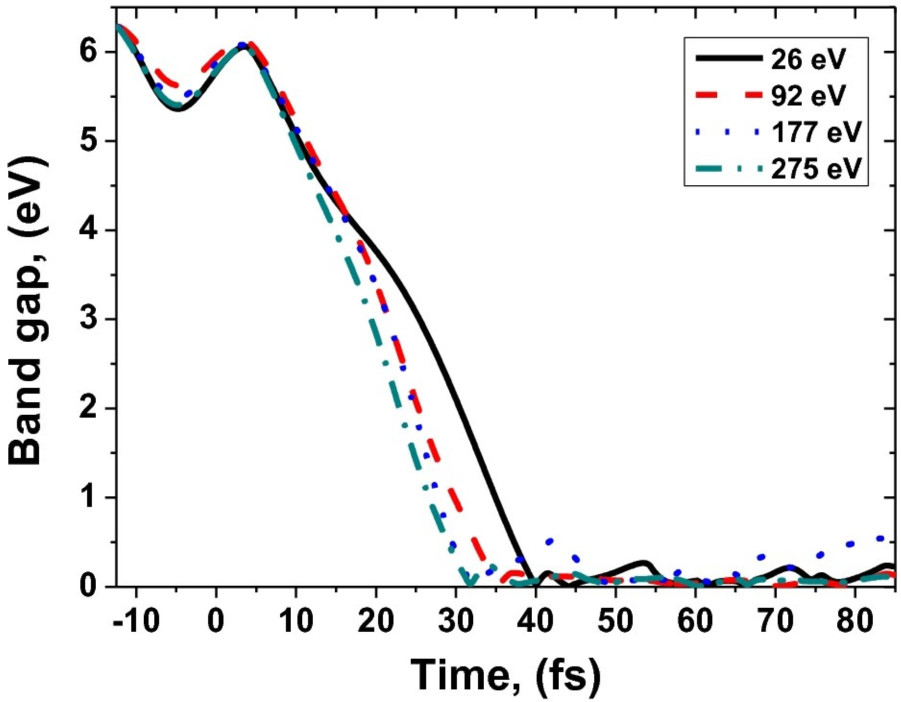
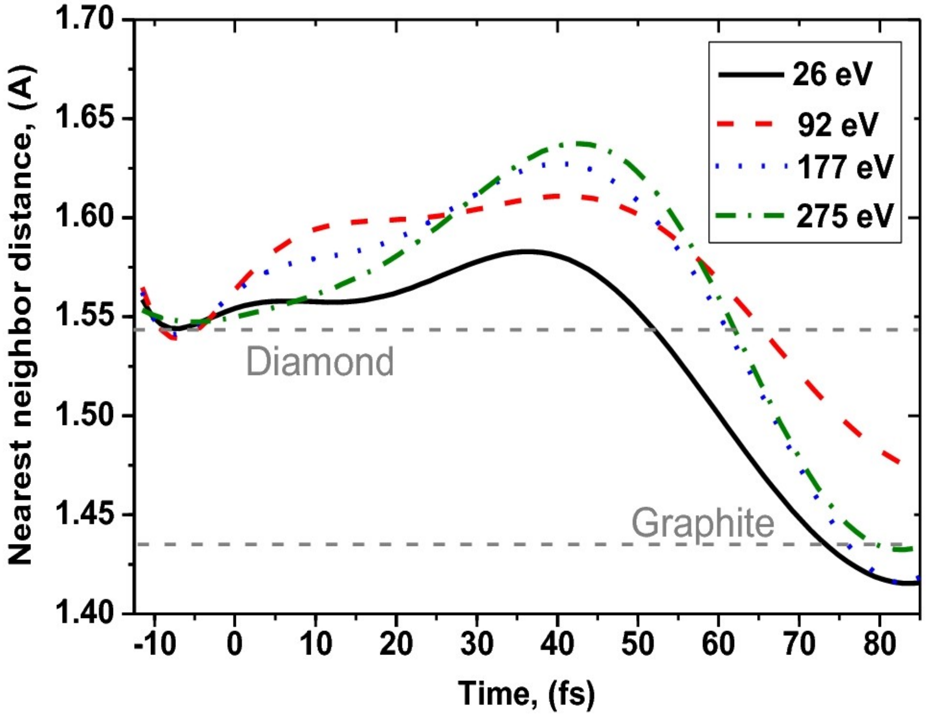
Results: Bandgap collapse

Different photon energies: 26 eV, 92 eV, 177 eV, 275 eV





Bandgap collapse induces ultrafast phase transition

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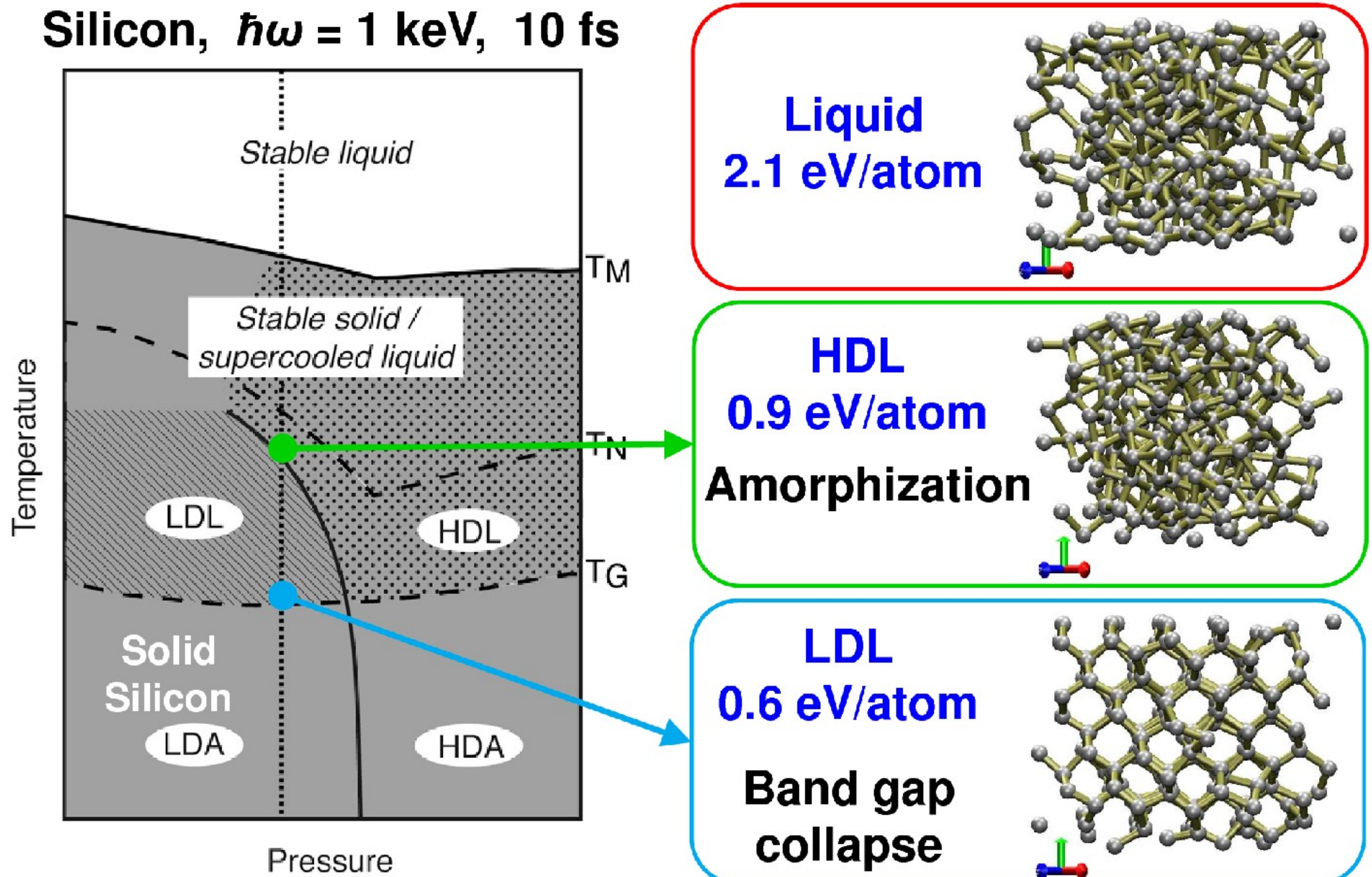
Structural transition in Si: interplay of thermal and non-thermal processes



M. Beye, *et al.*, *J. of El. Spectr.* 188 (2013) 172

N. Medvedev, Z. Li, B. Ziaja, PRB 91 (2015) 054113

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Diagnostics of transitions?

Damage thresholds → post mortem measurements on samples

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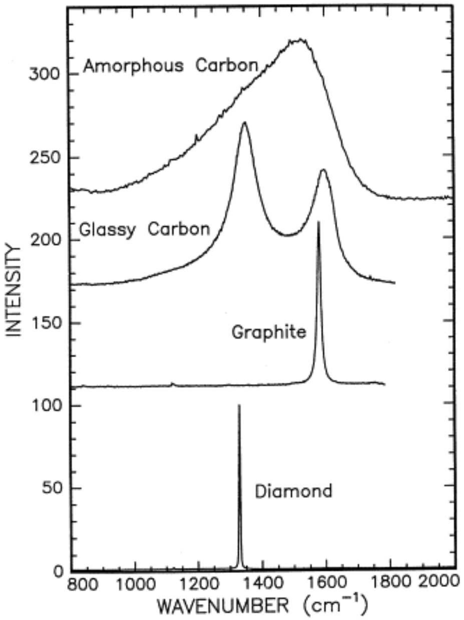


matsci4uwi.wordpress.com

Time-resolved diagnostics of transitions:

Pump-probe experiments:

- pump pulse initiates transition ...

- probe pulse probes it at varying time delay ...

Ultrashort pulses from XFELs enable this!

Transient optical properties as diagnostics of structural transitions within irradiated systems

Low material excitation below and around damage threshold → band structure evolution accurately described with transferable tight binding method

[V. Tkachenko, N. Mevedev et al.(BZ), PRB, in revision]

Long-wavelength limit (q → 0), **Tight-binding (TB) model**

Optical **dielectric function** within the random-phase approximation (Lindhard formula) [3]:

$$\epsilon^{\alpha\beta}(E) = \delta_{\alpha,\beta} + \frac{4\pi e^2\hbar^2}{m\Omega}\sum_{n,n'}(\eta_{n'} - \eta_n)\frac{F^{\alpha\beta}_{n,n'}}{E_{n,n'}}\left[\frac{1}{E - E_{n,n'} + i\gamma}\right]$$

$$F^{\alpha\beta}_{n,n'} = \frac{2\langle n|\hat{p}_\alpha|n'\rangle\langle n'|\hat{p}_\beta|n\rangle}{mE_{n,n'}}$$ - the oscillator strength [3]

Calculated within tight-binding model by F. Trani et al, as: $\mathbf{P}(\mathbf{R},\mathbf{R}') = \frac{m}{i\hbar}[\mathbf{R} - \mathbf{R}']H(\mathbf{R},\mathbf{R}')$

Dielectric function → refractive indices n, k

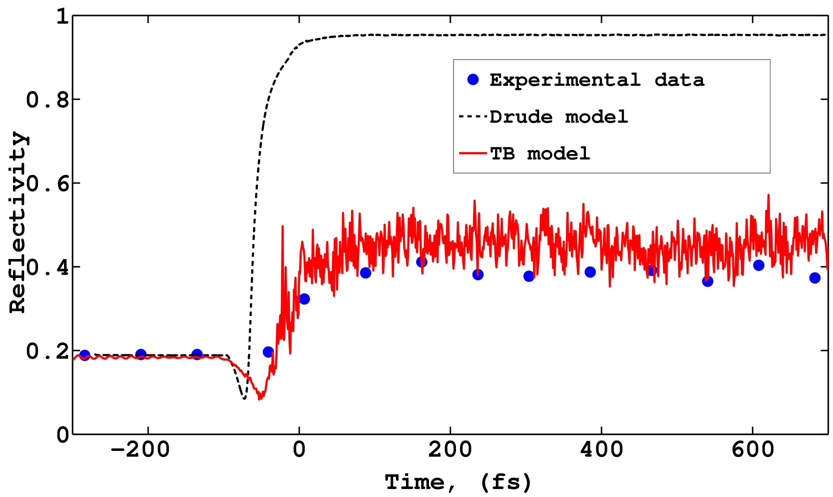
[Courtesy of V. Tkachenko]

Transient optical properties as diagnostics of structural transitions within irradiated systems

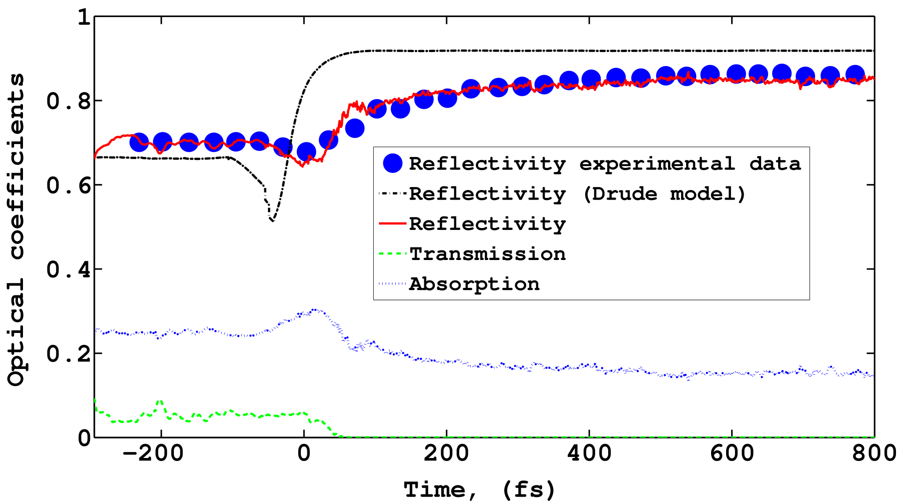
Low material excitation
below and around damage threshold → band structure evolution accurately described with transferable tight binding method

[V. Tkachenko, N. Medvedev et al.(BZ), PRB, in revision]

-Diamond and silicon are excited with a laser pulse ...
-Transient optical properties are probed with the optical laser pulse ...
-Complex dielectric function calculated from an ab-initio scheme ...



Diamond



Silicon

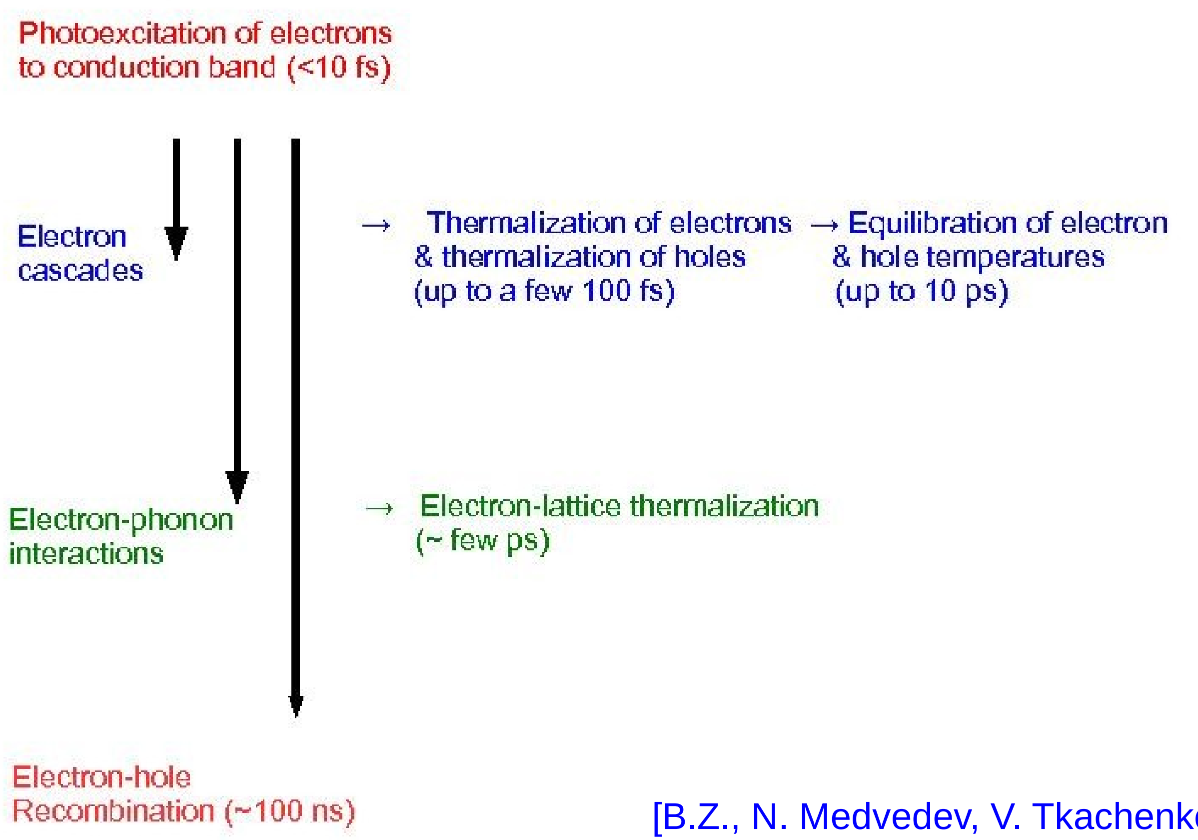
[Courtesy of V. Tkachenko]

Transient optical properties as diagnostics of structural transitions within irradiated systems

Low dose

Reflectivity overshooting in GaAs ← effect of band gap shrinking

Timescales of relaxation and excitation processes.



Damage Threshold

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[Courtesy of V. Tkachenko]

[B.Z., N. Medvedev, V. Tkachenko, T. Maltezopoulos, W. Wurth, *Sci. Rep*. **5**, 18068 (2015)]

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Transient optical properties as diagnostics of structural transitions within irradiated systems

Low dose

Reflectivity overshooting in GaAs

- Rate equations → the evolution of free-carrier densities as a function of time [5];

$$d\,n_{e-h}(t)/dt \;=\; \gamma_{e-h}(t)$$ ← Before ΔR/R minimum

$$d\,n_{e-h}(t)/dt = -\gamma_{rec}\cdot n_{e-h}(t)$$ ← After ΔR/R minimum

- Two-temperature model → electron-lattice equilibration [5];

$$d\,T_{latt}(t)/dt \;=\; +G_{latt}(T_{e-h}(t) - T_{latt}(t))$$

$$d\,T_{e-h}(t)/dt \;=\; -G_{e-h}(T_{e-h}(t) - T_{latt}(t))$$

- Drude model → follows the transient reflectivity (extended for interband transitions) [5].

Damage Threshold

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[Courtesy of V. Tkachenko]

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[B.Z., N. Medvedev, V. Tkachenko, T. Maltezopoulos, W. Wurth, *Sci. Rep*. **5**, 18068 (2015)]

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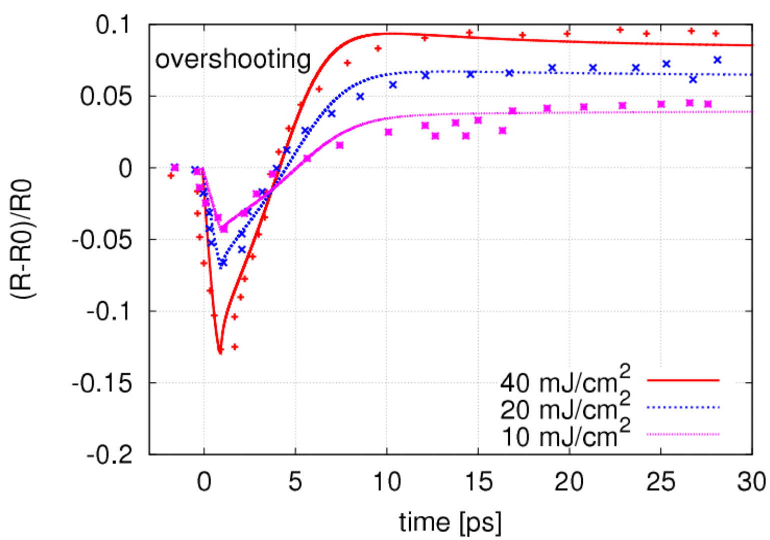
Transient optical properties as diagnostics of structural transitions within irradiated systems

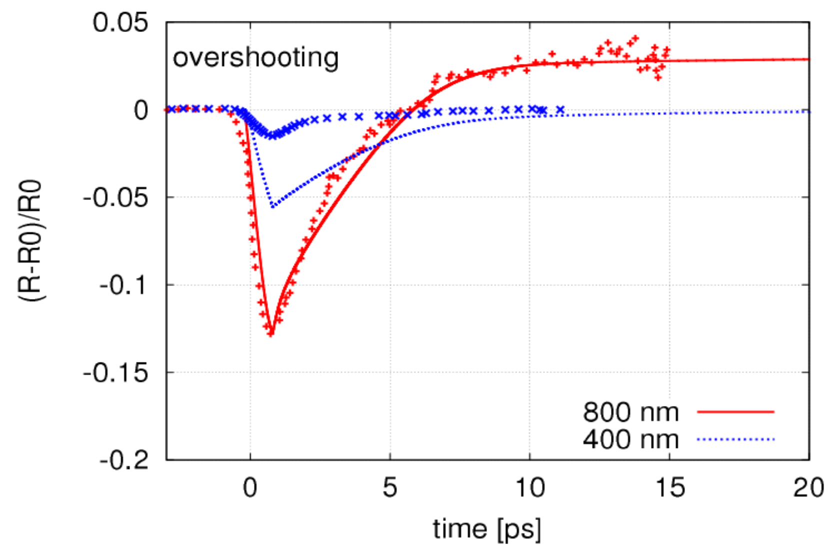
Low dose

Reflectivity overshooting in GaAs ← effect of band gap shrinking

- Timescale of a few ps
- Observable at probe wavelength ~ band gap width (low absorption)
- Measurement of electron-phonon coupling with femtosecond resolution ($\tau_{el\text{-}latt}$~ 2-3 ps) and transient electronic temperatures (~ 2-3 eV)
- Expected for other narrow band-gap semiconductors



LCLS measurement (800 eV)



FLASH measurement (40 eV)

Damage Threshold

[B.Z., N. Medvedev, V. Tkachenko, T. Maltezopoulos, W. Wurth, *Sci. Rep*. **5**, 18068 (2015)]

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[Courtesy of V. Tkachenko]

Transient optical properties as diagnostics of warm dense matter formation

Prospects:

Tight-binding method can be used only to low-excited materials ...

How about transition to warm dense matter after high material excitation?

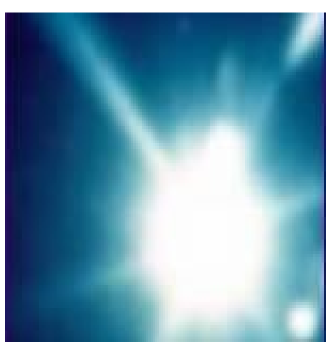
↓

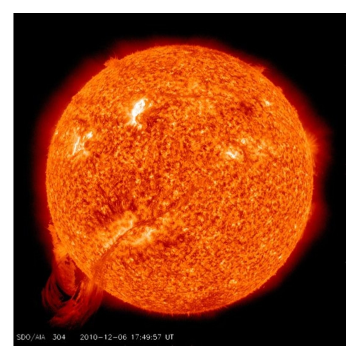
Dedicated ab-initio method needed to describe non-equilibrium changes within band structure ← adapted from XMOLECULE scheme?

↓

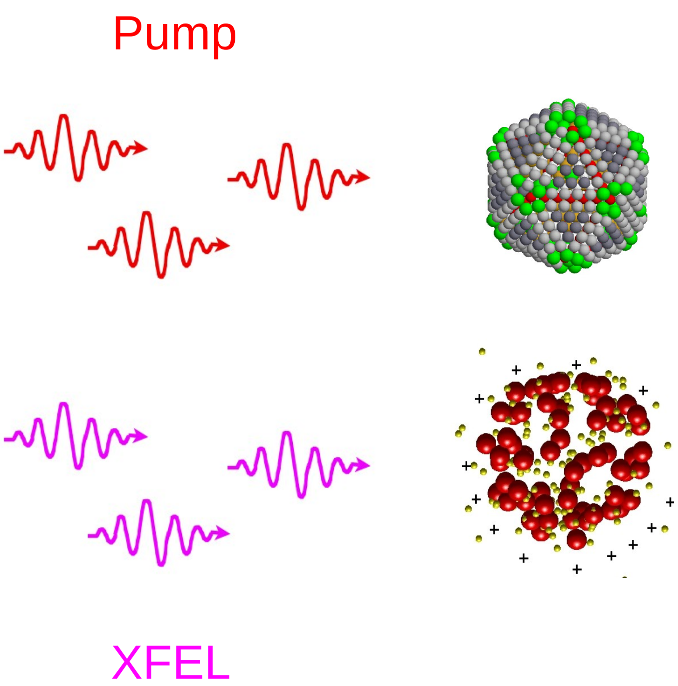
[Y. Hao et al., Str. Dyn. 2 (2015) 041707]

Transition can be monitored with transient optical properties ...





X-ray diffraction as diagnostics of structural transitions within irradiated systems



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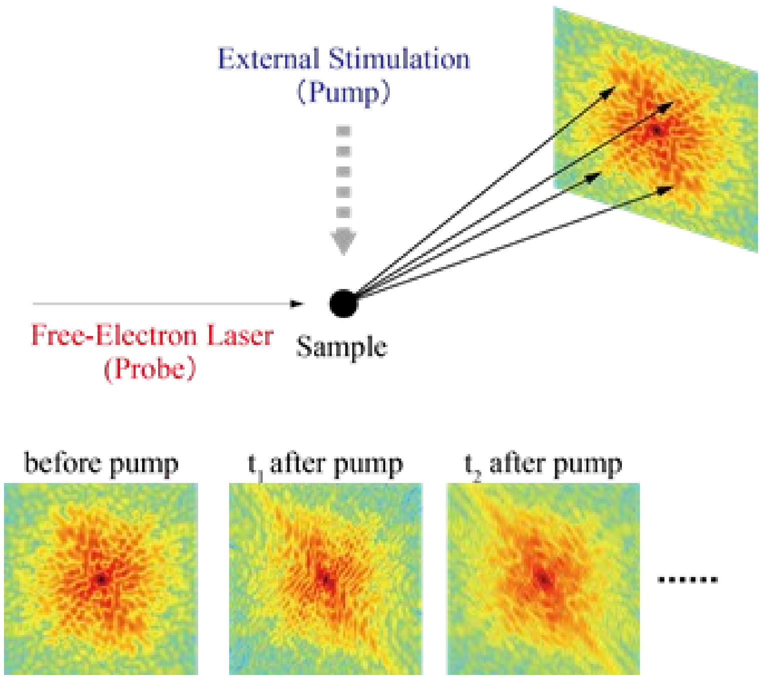


[cxo-www.es.hokudai.ac.jp]

Summary

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

-below damage threshold – non-equilibrium electron kinetics

-below melting threshold – also rearrangement of atomic structure

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

Time-resolved diagnostics of transitions

- transient optical properties

- X-ray diffraction

- ...

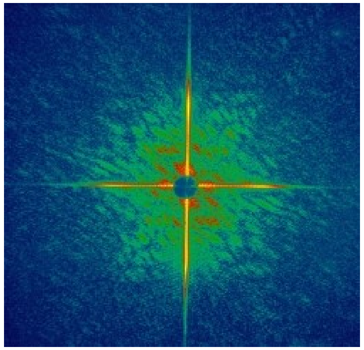
Thanking my collaborators and CFEL Theory Division

N. Medvedev

V. Tkachenko

Thanking our external collaborators...

J. Gaudin (CELIA, Bordeaux)



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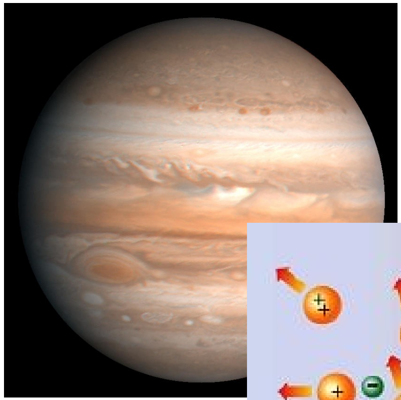
H.-K. Chung (IAEA, Vienna), R. W. Lee (LBNL, Berkley)



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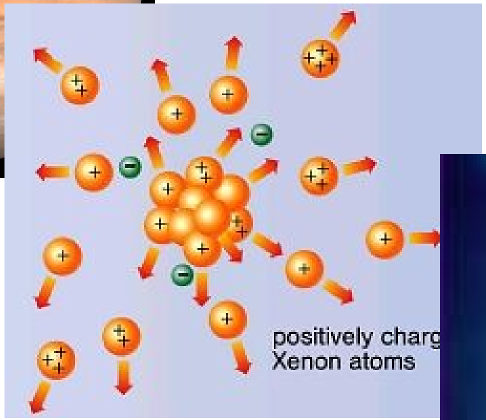
T. Malezopoulos (U. Hamburg)

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



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

W.Wurth (DESY)

and ...

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Summary of codes

Atomic data and transitions rates from XATOM package (R. Santra & S. K. Son)

Transport approach → Boltzmann code: irradiation of atomic clusters with VUV and soft X-ray photons (B. Ziaja) → recently also extended to hard X-ray regime

Hybrid MC-TBMD code XTANT → modeling of structural changes in irradiated solids → applied to describe nonthermal melting in semiconductors (N. Medvedev) → transient optical properties (V. Tkachenko)

Monte Carlo model to follow ultrafast electron kinetics in X-rays irradiated solids (N. Medvedev); an element-flexible fast version XCASCADE → transient optical properties beyond Drude model

Molecular Dynamics code to follow classical ion and electron dynamics in X-ray irradiated clusters and macromolecules (Z. Jurek)→ XHYDRO: its long-timescale extension (V. Saxena); on-the-fly coupling to XATOM working (S. K. Son)

Thank you for your attention !