Thermal and non-thermal transitions in X-ray-irradiated solids

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Outline

1. Introduction
2. Model: XTANT
3. Nonthermal phase transitions
4. Thermal phase transitions
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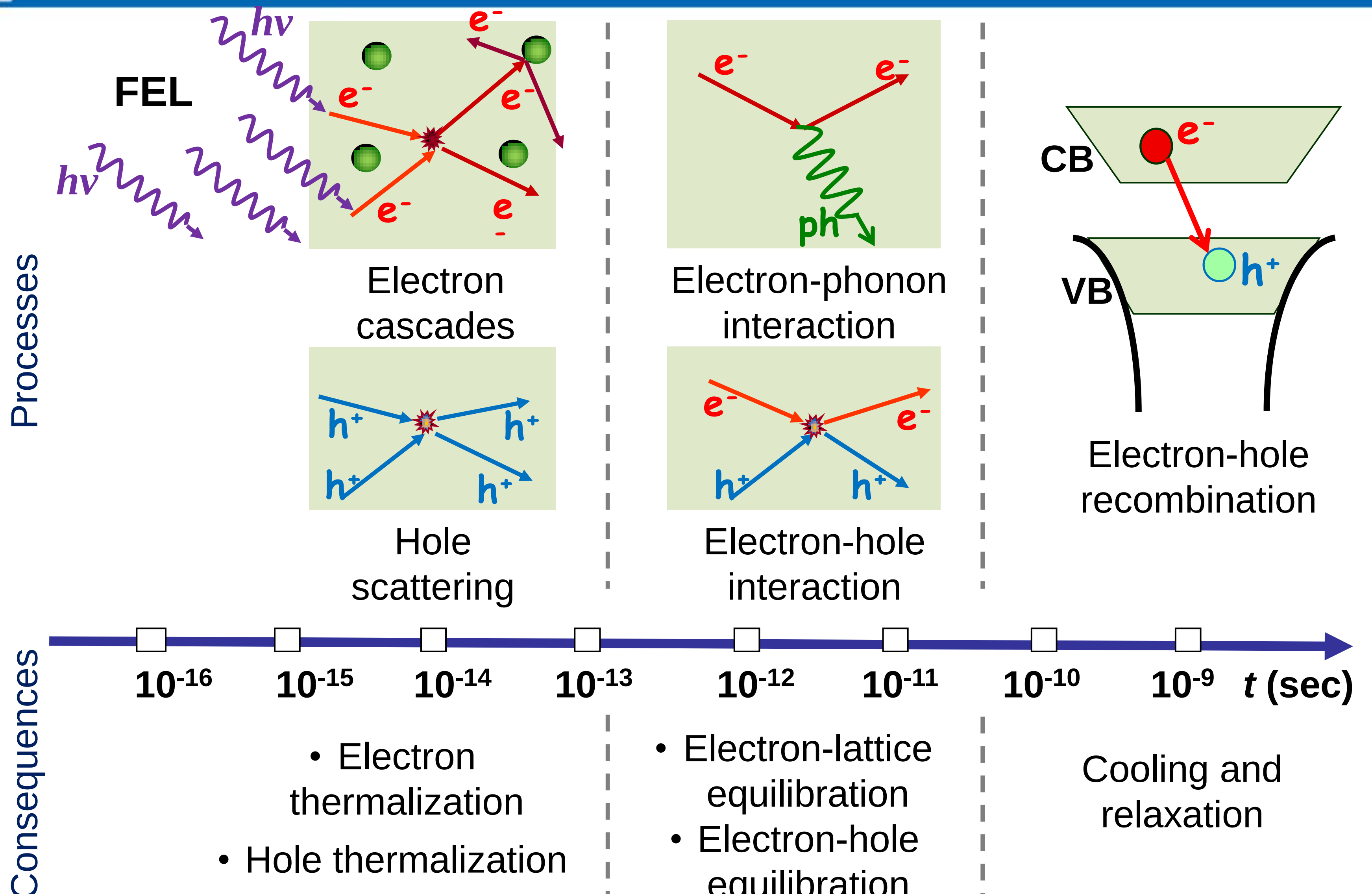
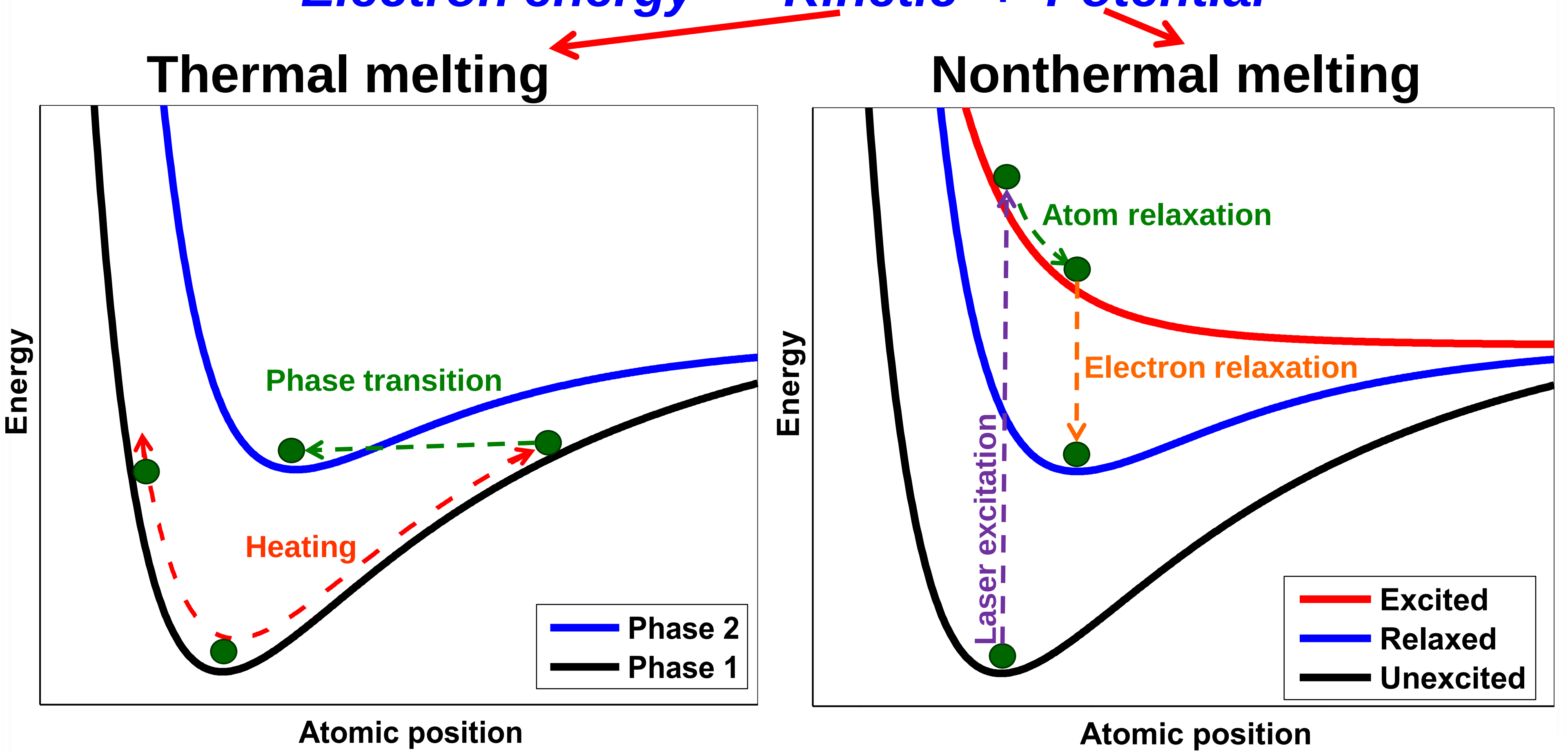
Introduction: timescales



[B. Ziaja, N. Medvedev *et al.*, Sci Rep. 5 (2015) 18068]

Thermal vs nonthermal

Electron energy = Kinetic + Potential



- **Thermal melting: atomic heating within the same potential**
- **Nonthermal melting: change of interatomic potential**

[N. Medvedev, A. Volkov, B. Ziaja, *NIMB* **365**, 437 (2015)]

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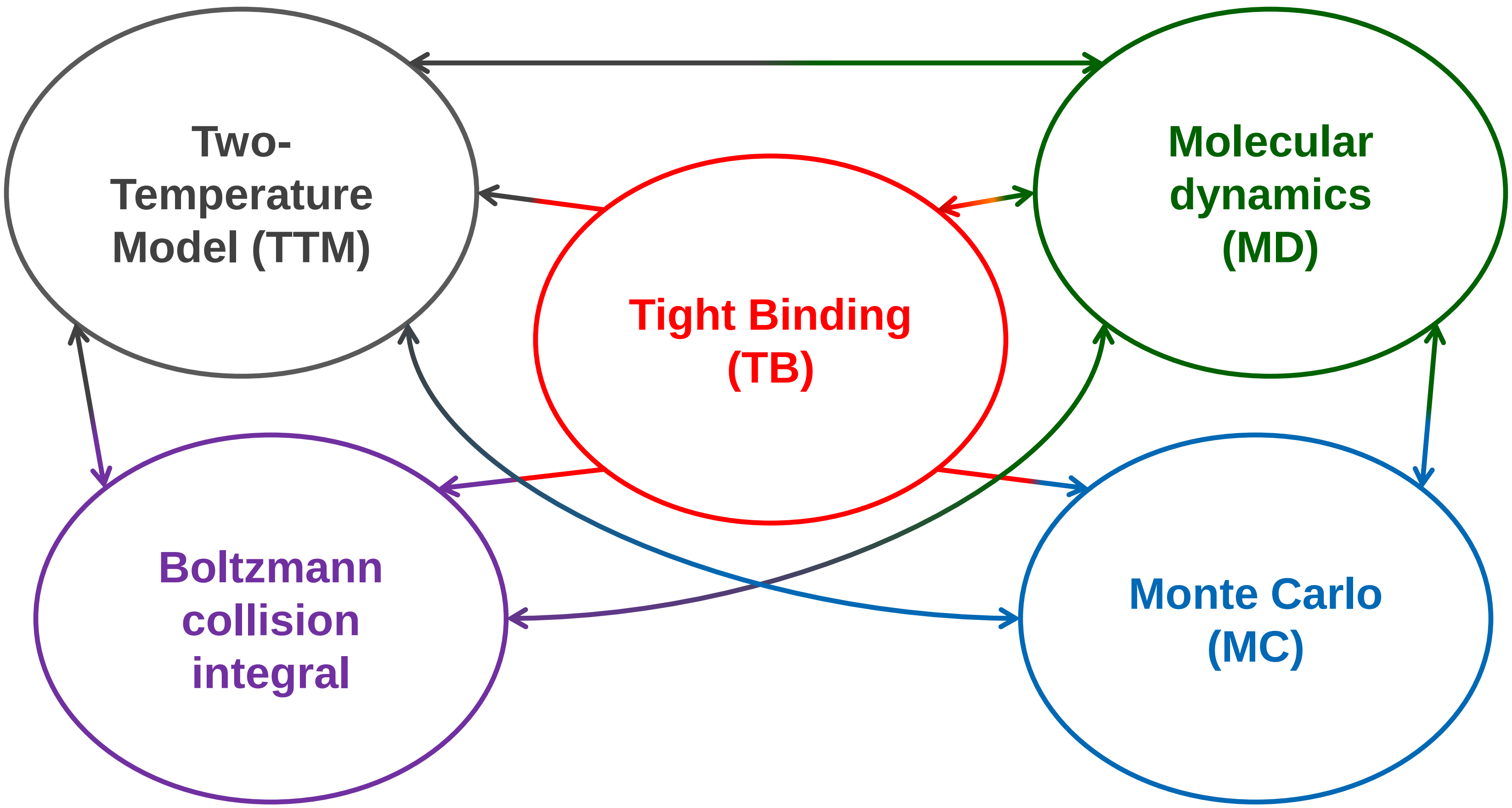
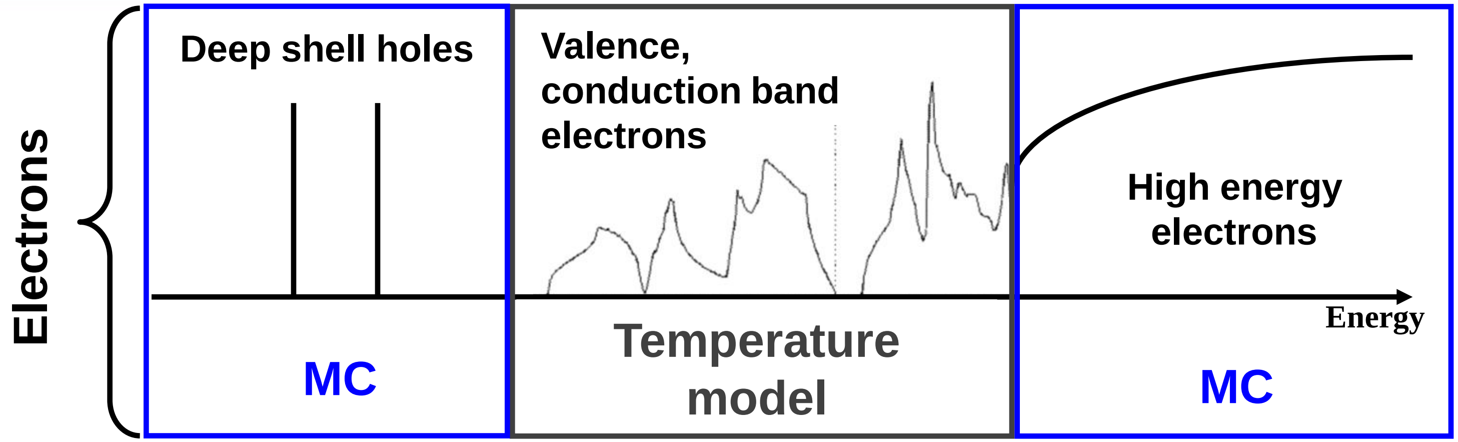
Hybrid model

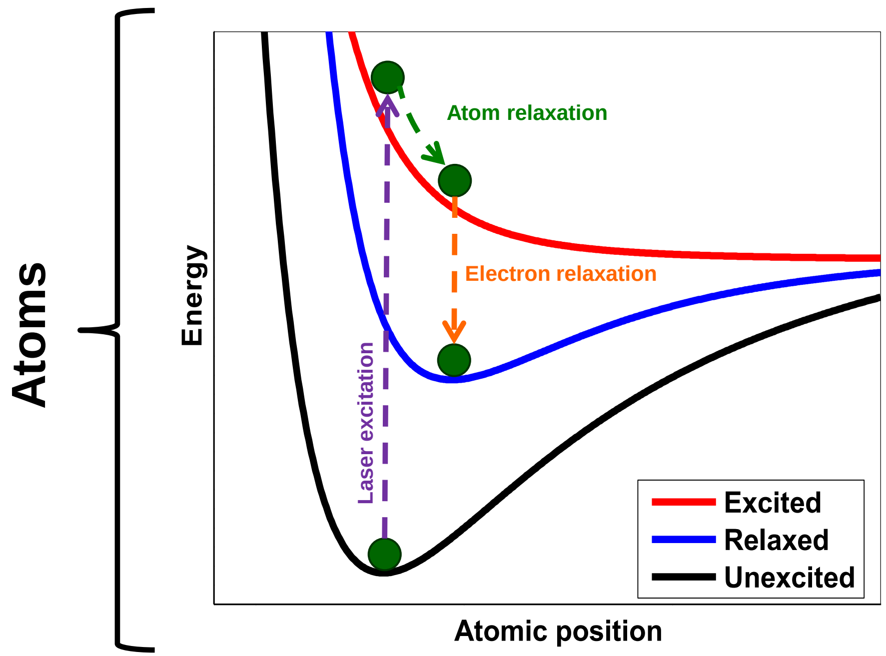
XTANT: X-ray-induced Thermal And Nonthermal Transitions

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



Schematics of XTANT





Tight Binding MD

+

Nonadiabatic energy exchange

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

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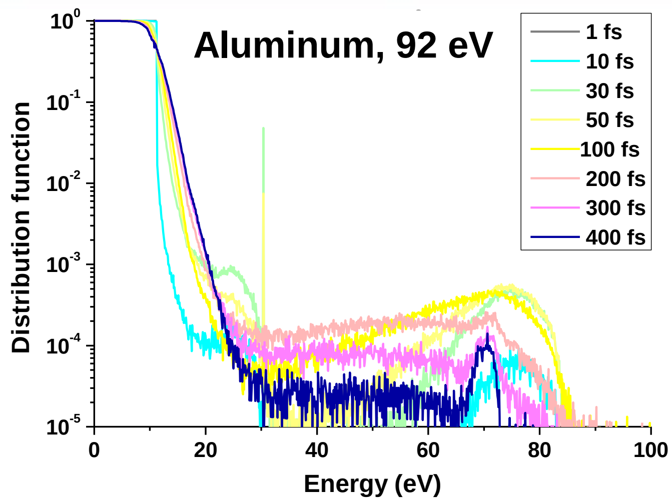
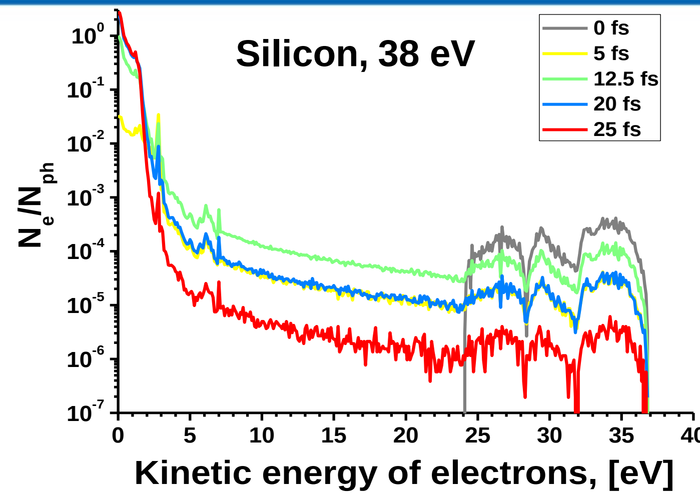
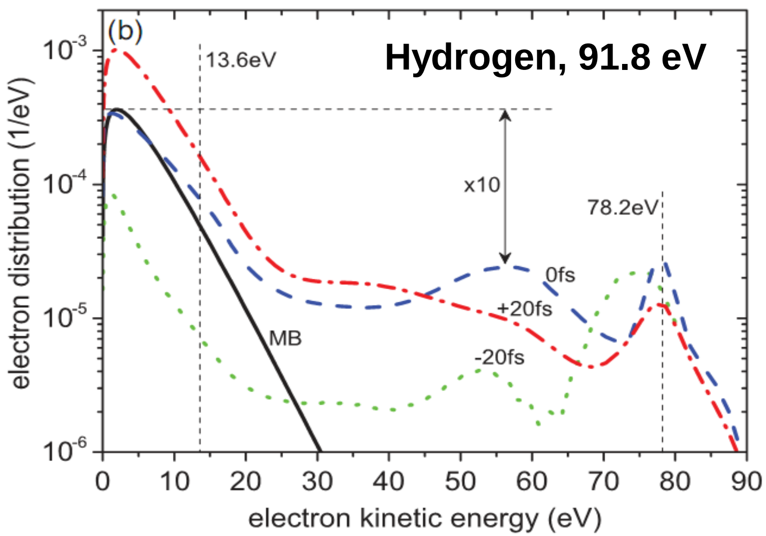
Bump-on-hot-tail distribution



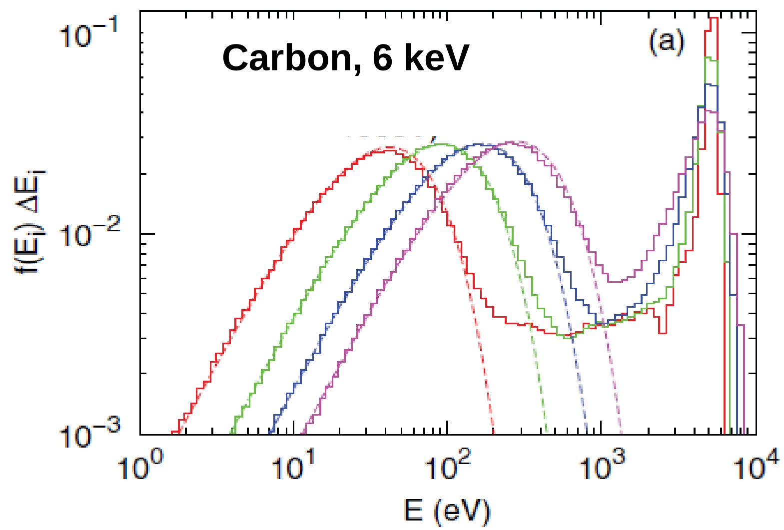
[N. Medvedev *et al.*, PRL **107** (2011)]



[N. Medvedev, B. Rethfeld, NJP **12** (2010)]



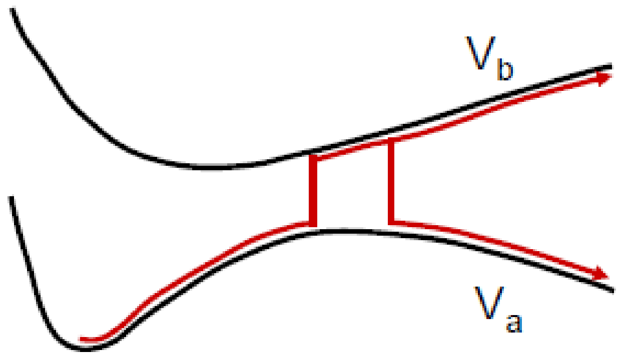
[R.R. Faustlin, B. Ziaja *et al.*, PRL **104** (2010)]



[S. P. Hau-Riege, PRE **87** (2013)]

Electron-ion coupling we use

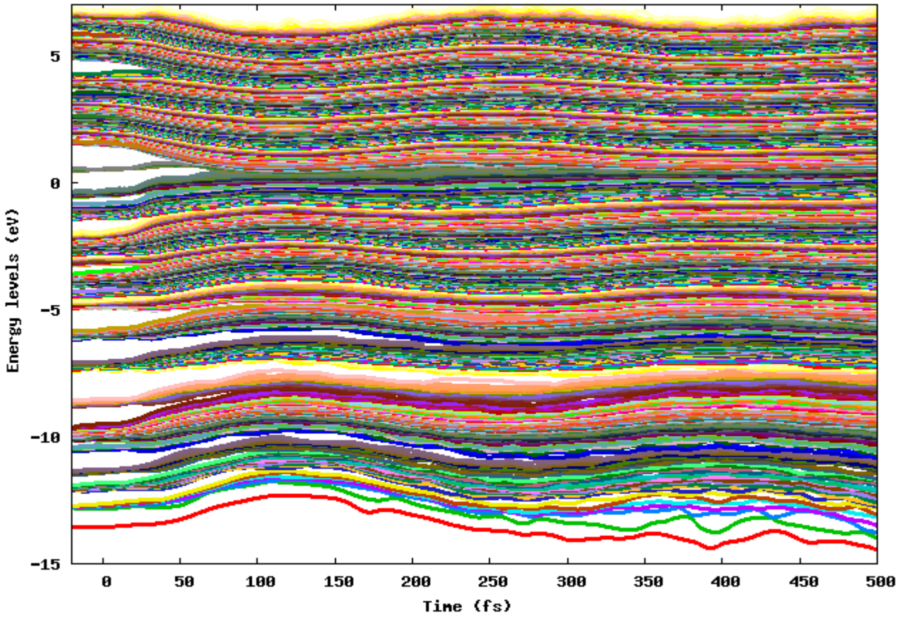
Femto-chemistry "surface-hopping method"



J. C. Tully, *J. Chem. Phys.* 93 (1990) 1061

Surface-Hopping

[http://simons.hec.utah.edu/TSTCsite/TullyLectures/]



N. Medvedev, *et al.*, *SPIE Proc.* (2015)

Hopping probability: matrix element => use in collision integral

$$\sum_{j=1}^{N} I_{i,j}^{e-at} = \frac{2\pi}{\hbar} \sum_{j=1}^{N} \int_0^{\infty} \int_0^{\infty} |M_{e-at}(E_i, E_j)|^2 \; F_{i,j}(E_i, E_j, E_{at}, E_{at}^{fin}) dE_{at} dE_{at}^{fin}$$

$$M_{e-at}(E_i, E_j) = \frac{1}{2} \left(\langle i(t-\delta t) | j(t) \rangle - \langle j(t-\delta t) | i(t) \rangle \right) \left(\overline{E_j} - \overline{E_i} \right)$$

[N. Medvedev *et al.*, Phys. Rev. B 95 (2017) 014309]

Processes considered in XTANT

1) **Photoabsorbtion by deep shells and VB**
2) **Scattering of fast electrons:**
   - **Deep shell ionizations**
   - **VB and CB scatterings**
3) **Auger-decays of deep holes**

} **- MC**

4) **Thermalization in VB and CB** — **- Temperature model**

Boltzmann eq.

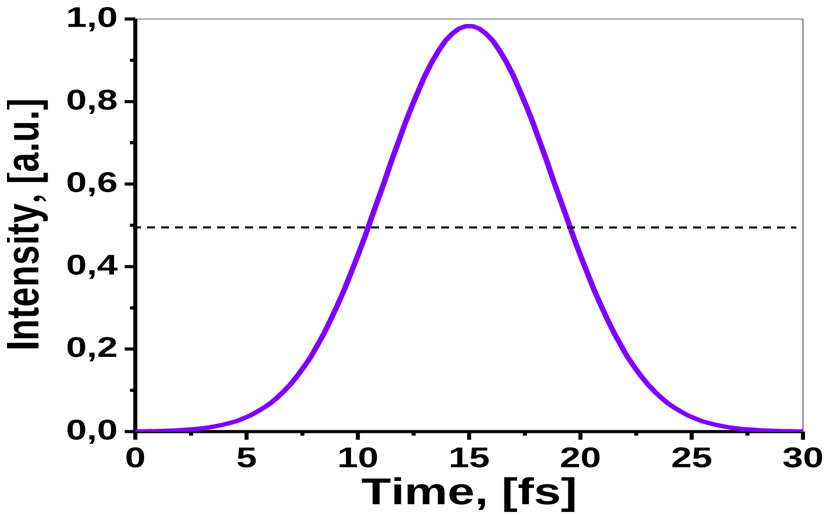
5) **Electron-ion coupling**
6) **Lattice heating, atomic dynamics**
7) **Changes of band structure**

} **- TBMD**

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

Outline

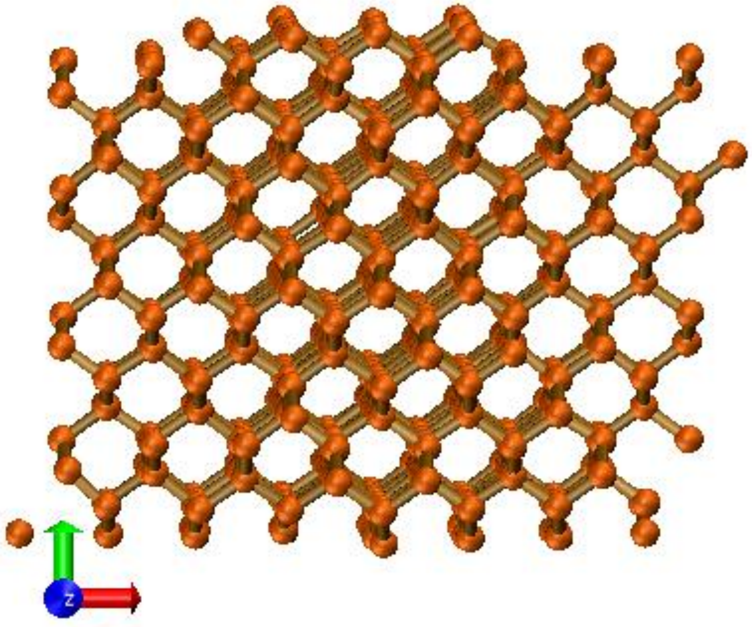
Parameters applied



Energy: ***hv* = 30 eV – 10 keV**

FWHM: **t_{pulse} ~ 10 fs**

Fluence: ***J* = 0.1 J/cm^2 – 10^3 J/cm^2**



Diamond target:

ρ = 3.5 g/cm^3

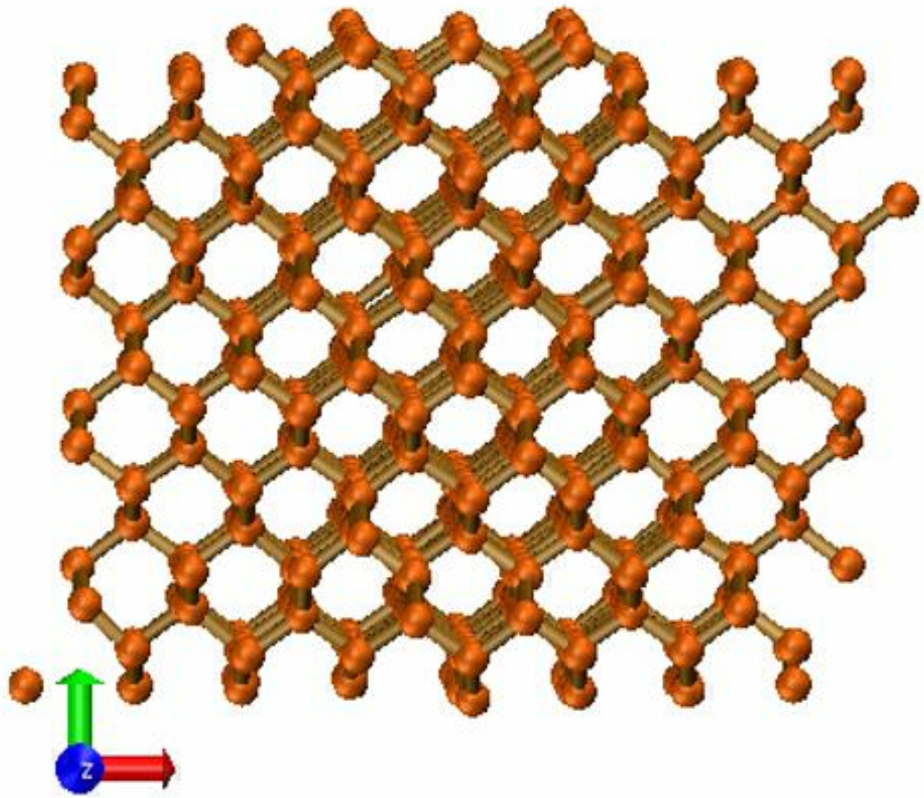
Silicon target:

ρ = 2.32 g/cm^3

216 - 1000 atoms

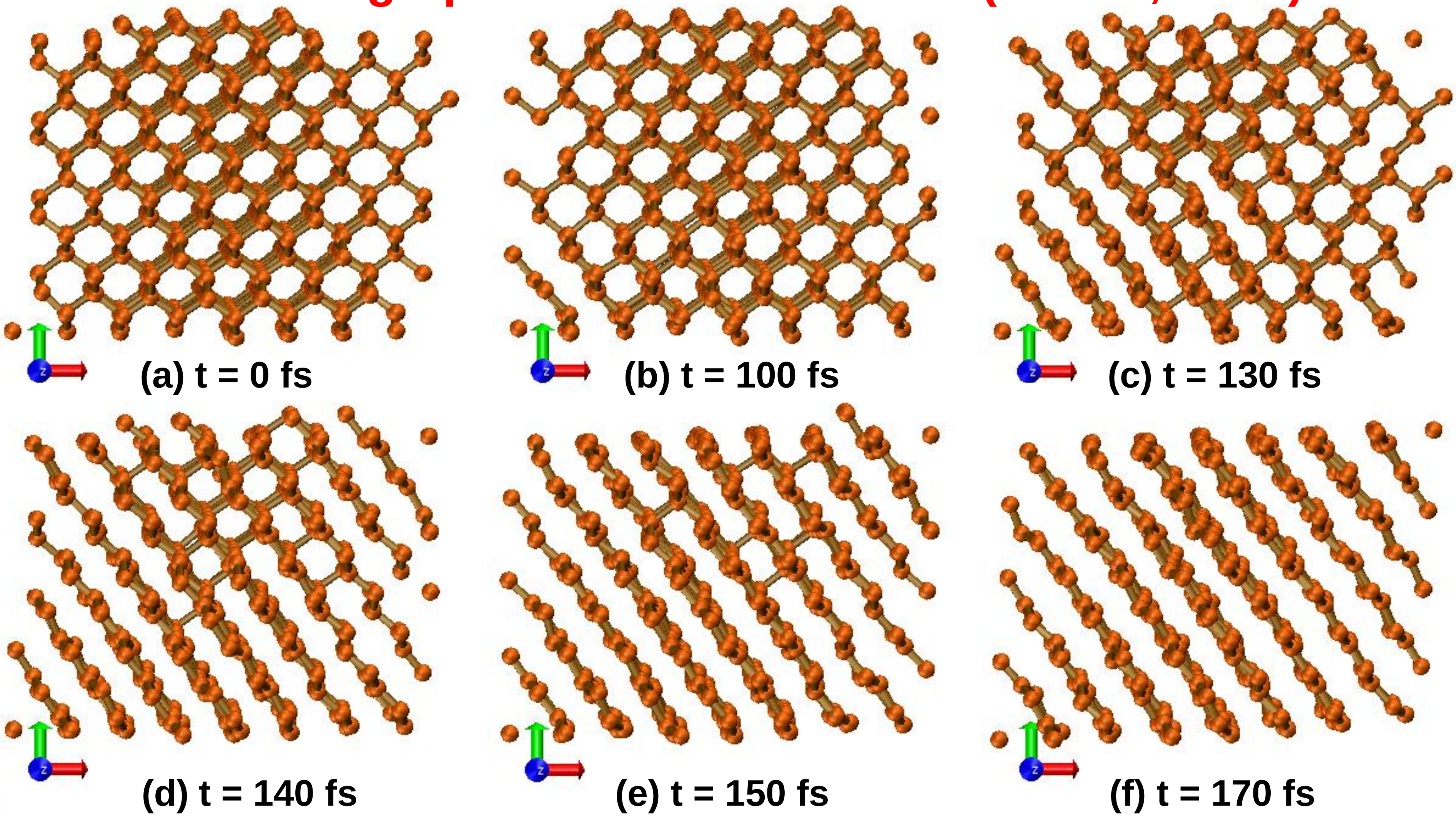
Nonthermal graphitization of diamond

$\hbar\omega$ = 10 keV, t = 10 fs, 0.85 eV/atom



Atomic snapshots

Ultrafast graphitization of diamond (10 keV, 10 fs)



[Medvedev, Tkachenko, Ziaja, Contrib. to Plasma Phys. 55 (2015) 12]

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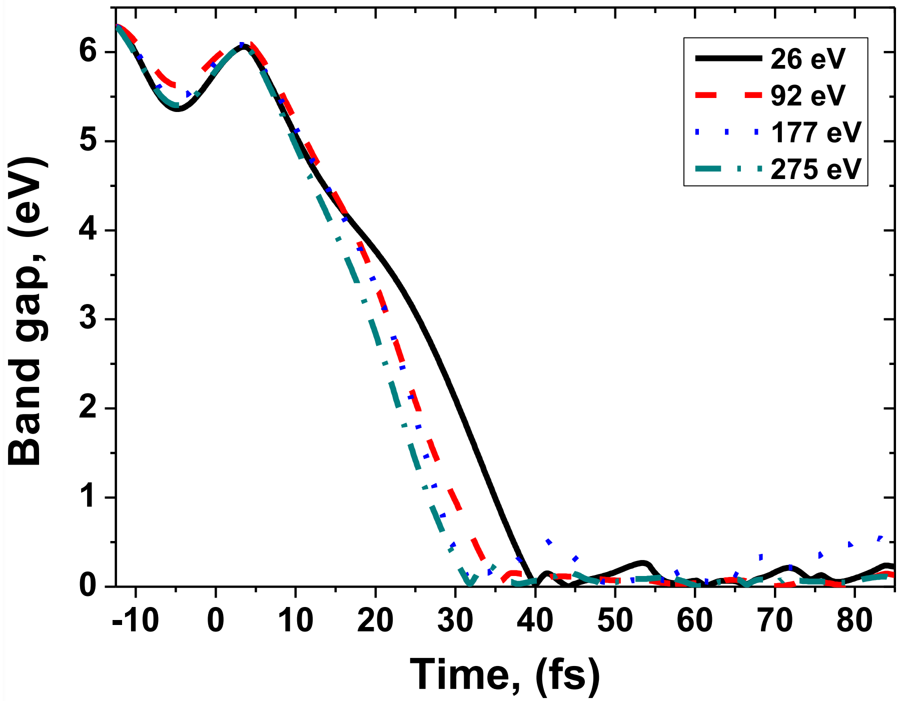
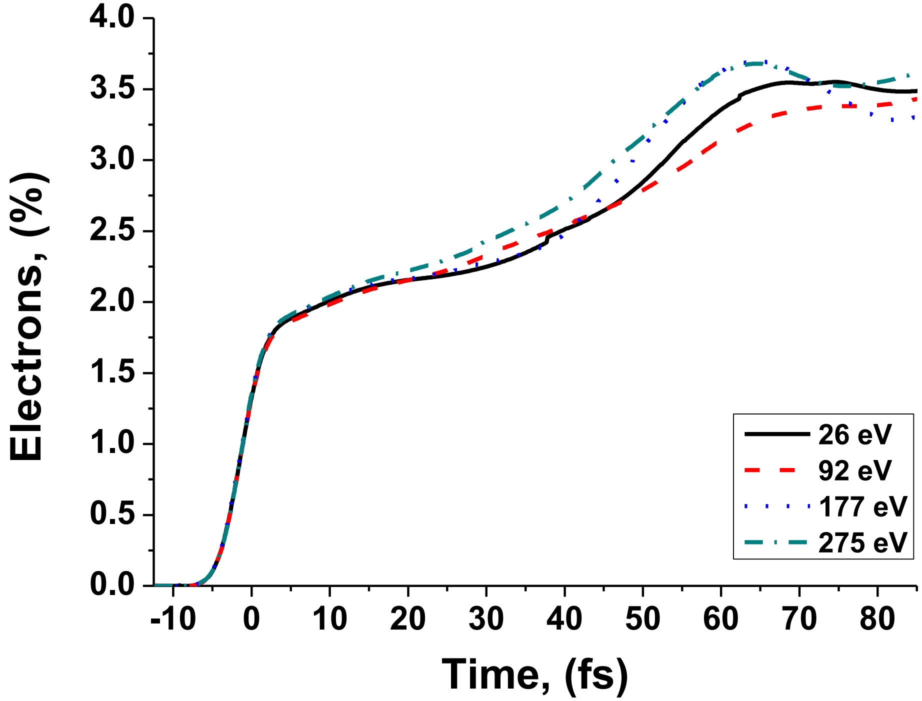
Band gap collapse

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





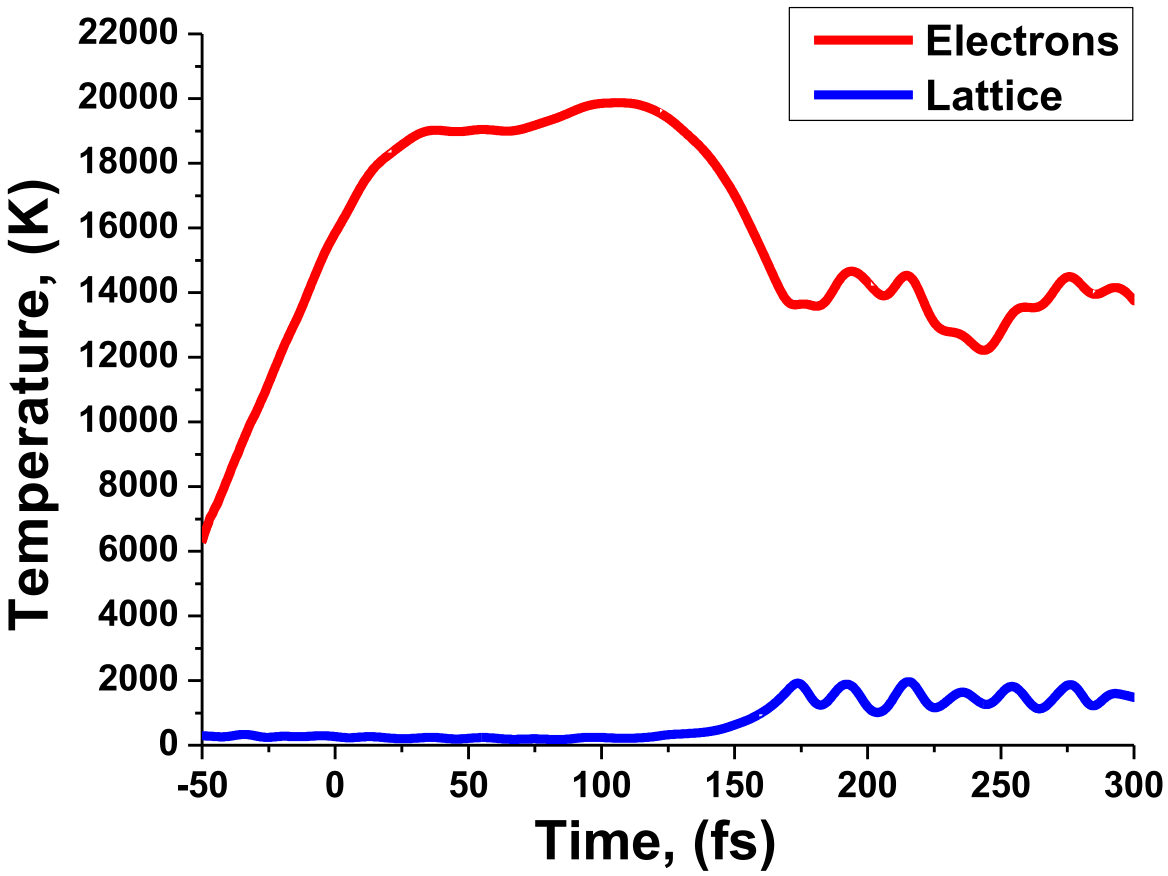
When electron density overcomes threshold value of 1.5 %, phase transition occurs (corresponds to dose 0.7 eV/atom)

Bandgap collapse induces ultrafast phase transition

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

Temperatures

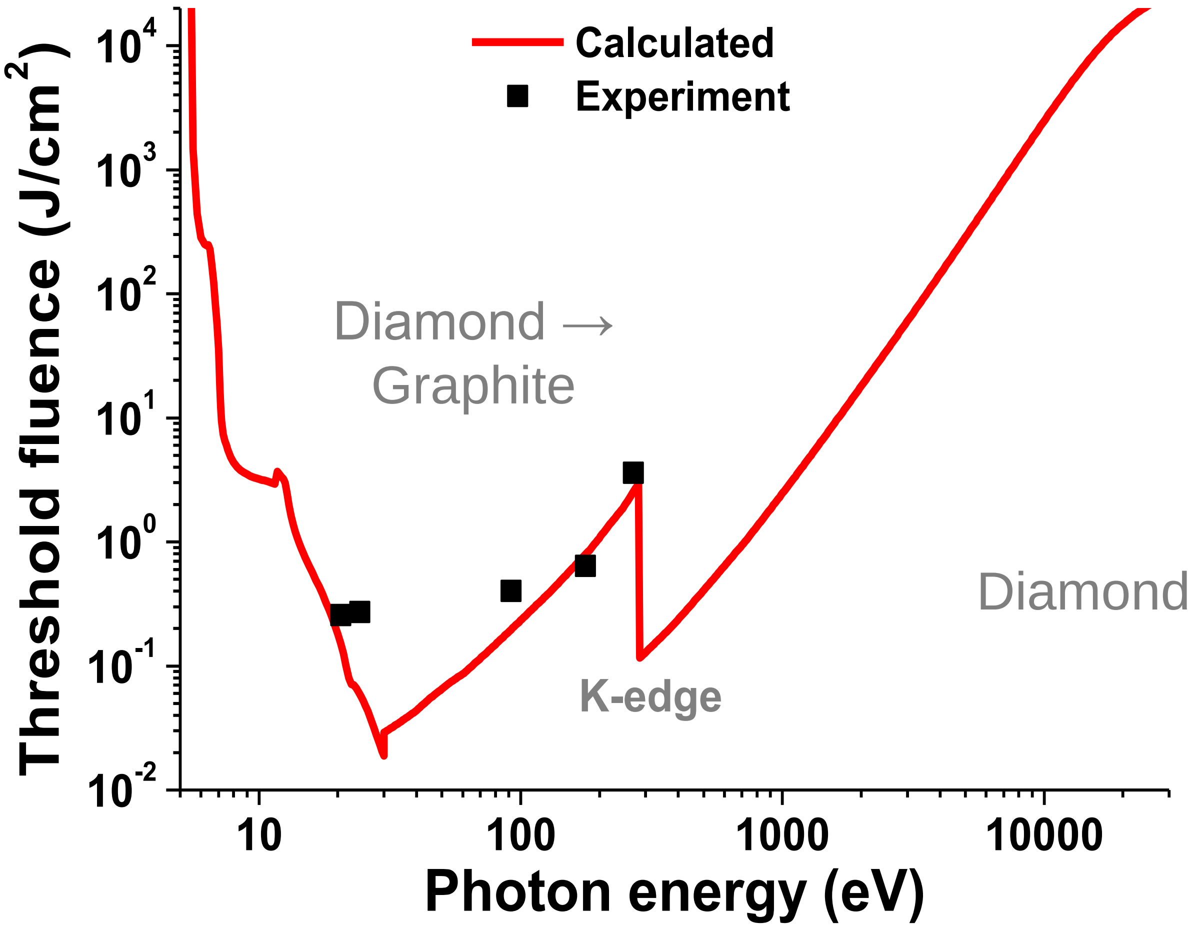
Photon energy 92 eV, FWHM = 50 fs, 0.85 eV/atom



Nonthermal phase transition:

Temperature increase follows *after* material modification, not induces it

X-ray damage threshold

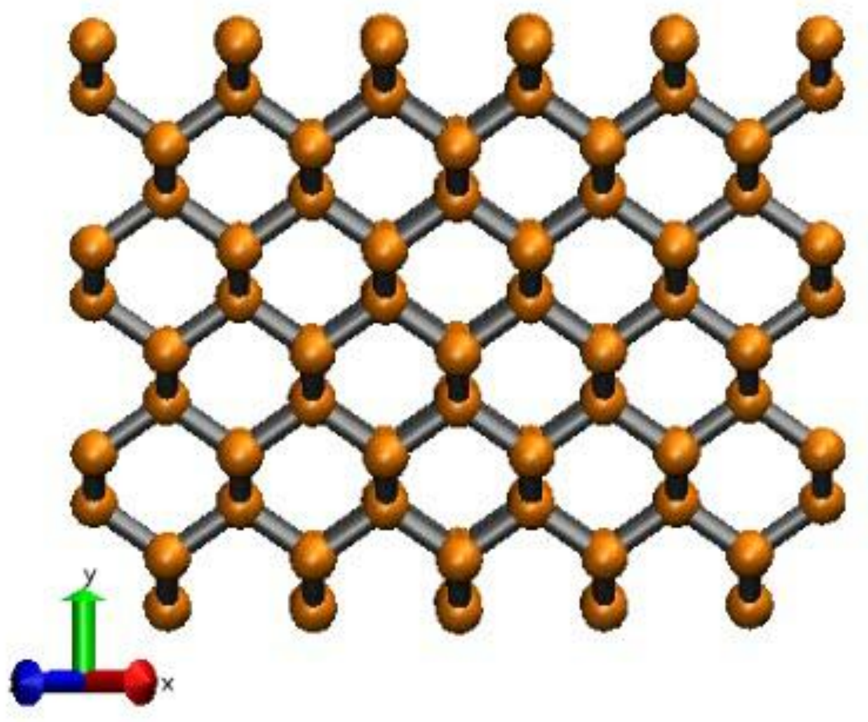


Damage threshold is in good agreement with experiments

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

Nonthermal melting of Si (HDL)

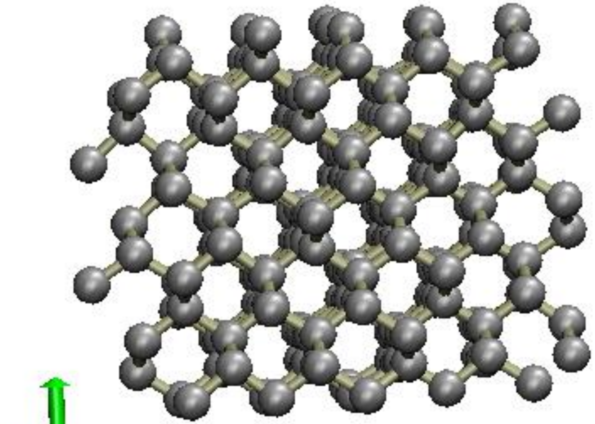
$\hbar\omega$ = 50 eV, FWHM = 10 fs, 1.1 eV/atom



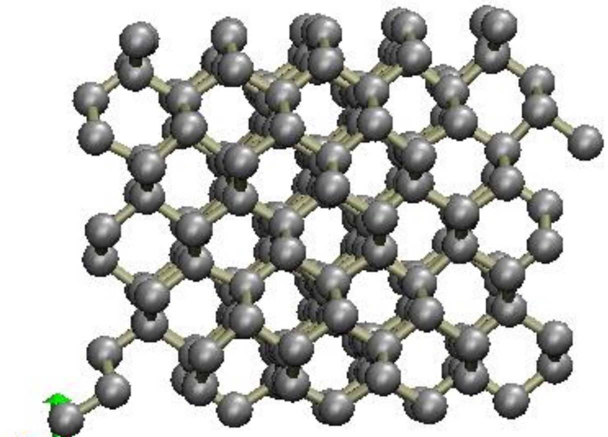
[N. Medvedev, Z. Li, B. Ziaja, Phys. Rev. B 91 (2015) 054113]

Snapshots of nonthermal melting

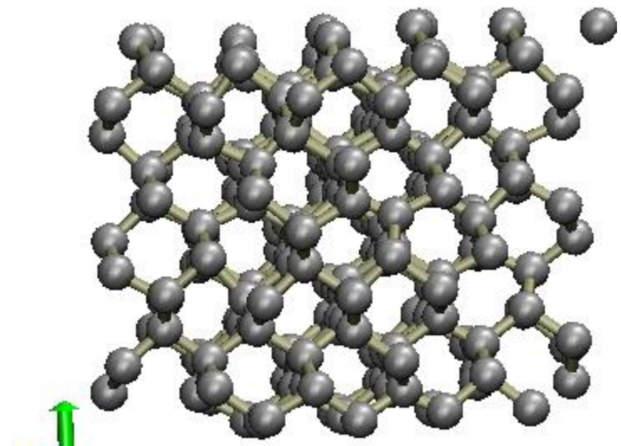
$\hbar\omega$ = 92 eV, FWHM = 10 fs, 2.5 eV/atom: 9% electrons



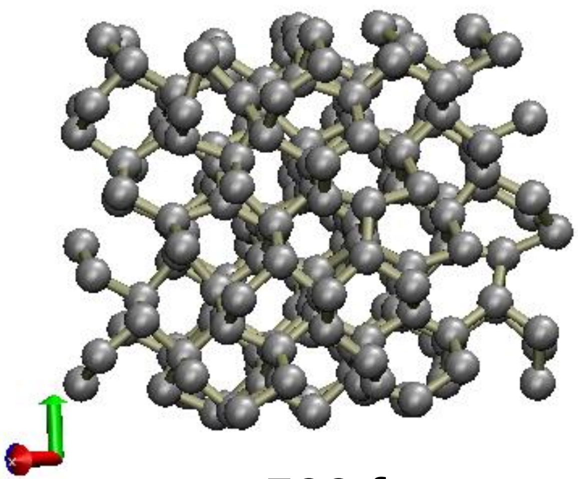
t = -100 fs



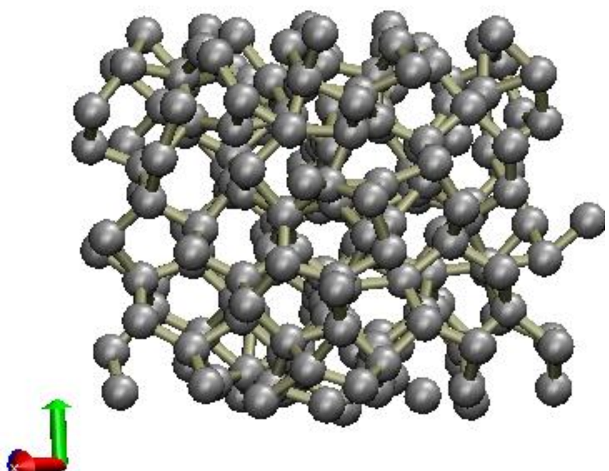
t = 0 fs



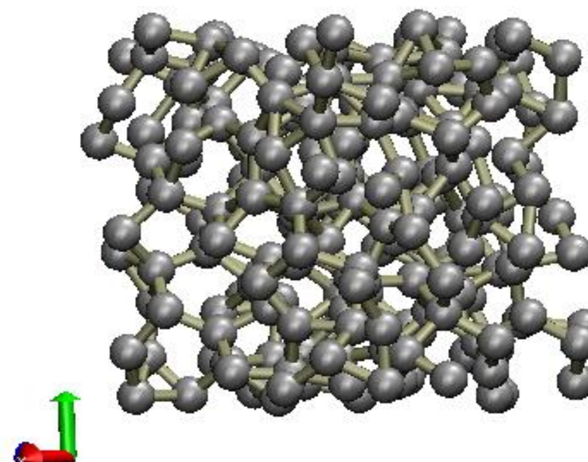
t = 300 fs



t = 500 fs



t = 800 fs



t = 1 ps

[N. Medvedev, Z. Li, B. Ziaja, Phys. Rev. B 91 (2015) 054113]

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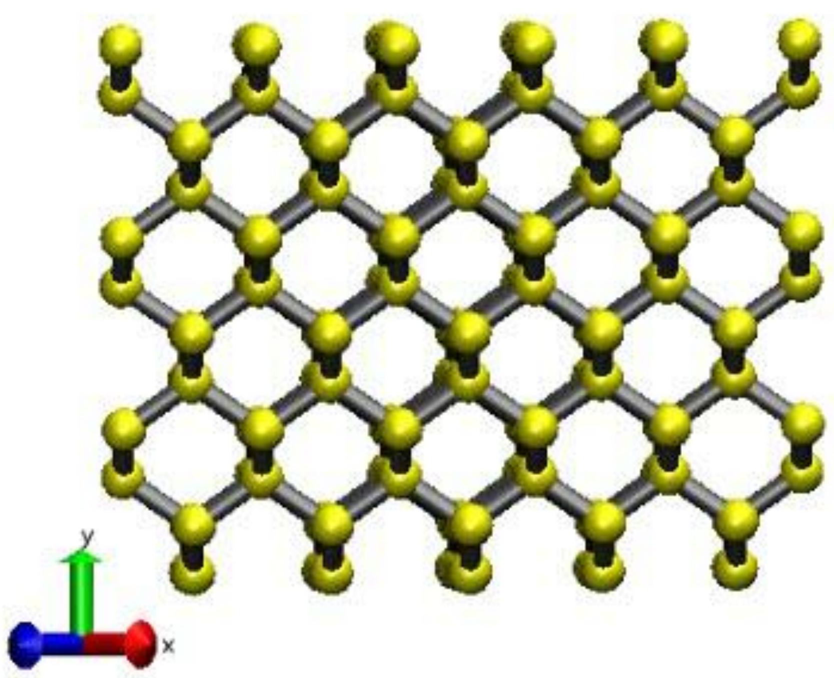
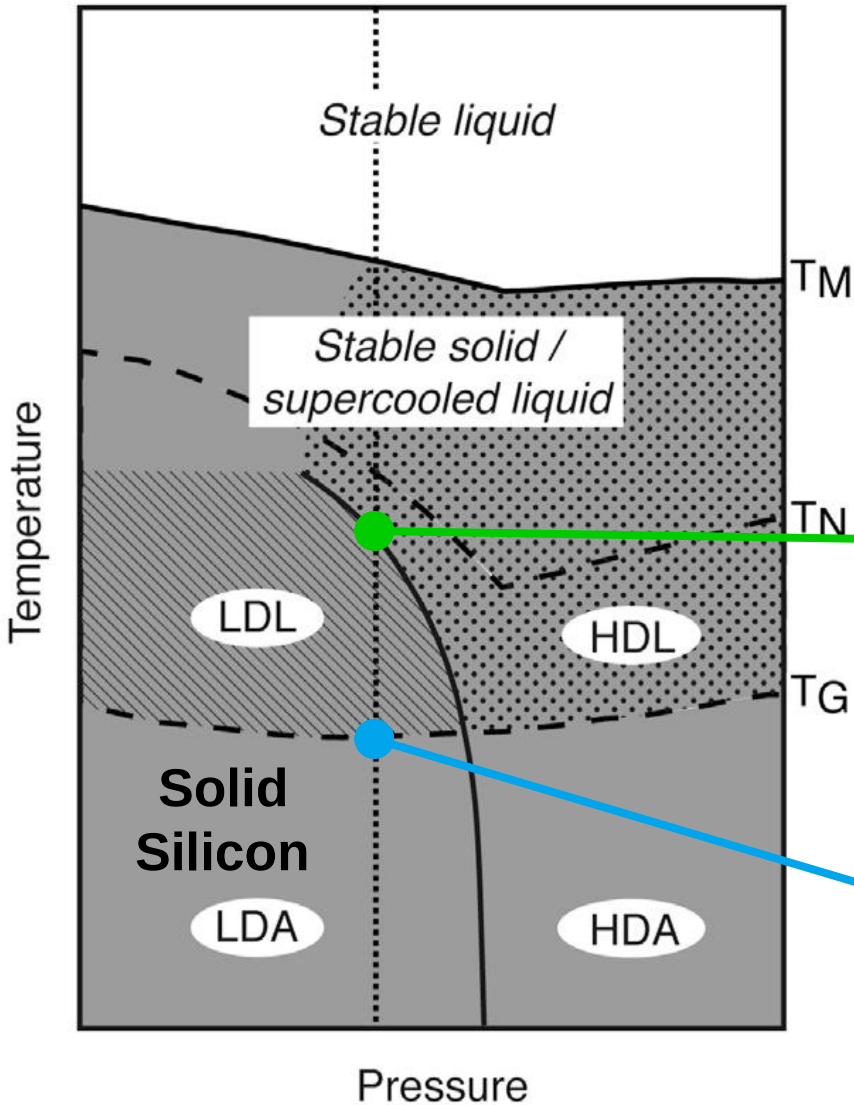
Thermal melting of Si (LDL)

$\hbar\omega$ = 50 eV, FWHM = 10 fs, 0.65 eV/atom

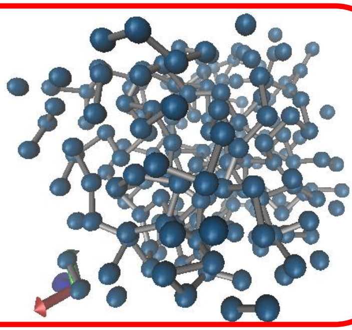


[N. Medvedev, Z. Li, B. Ziaja, Phys. Rev. B 91 (2015) 054113]

Thermal vs nonthermal effects

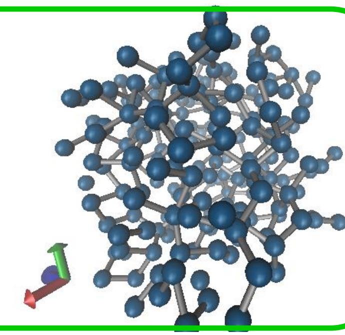


Liquid
(2.1 eV/atom;
T_e=17000 K)
9 % electrons



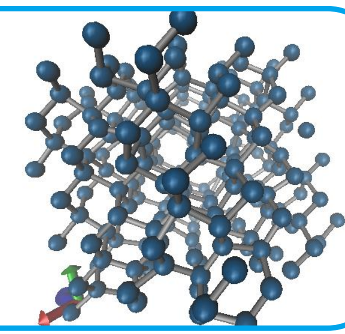
HDL
0.9 eV/atom

Amorphization
~5% electrons



LDL
0.6 eV/atom

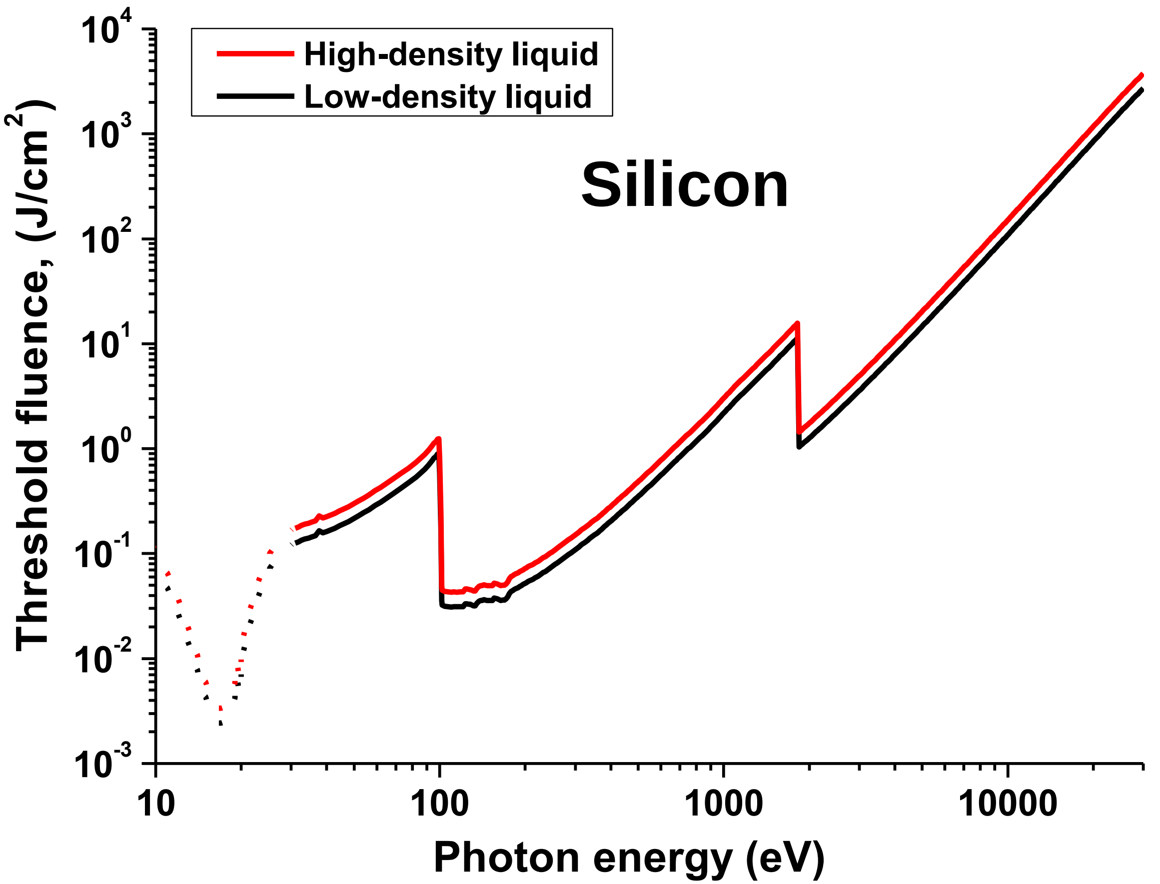
Band gap collapse



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

N. Medvedev, *et al*, PRB 91 (2015) 054113

X-ray damage threshold

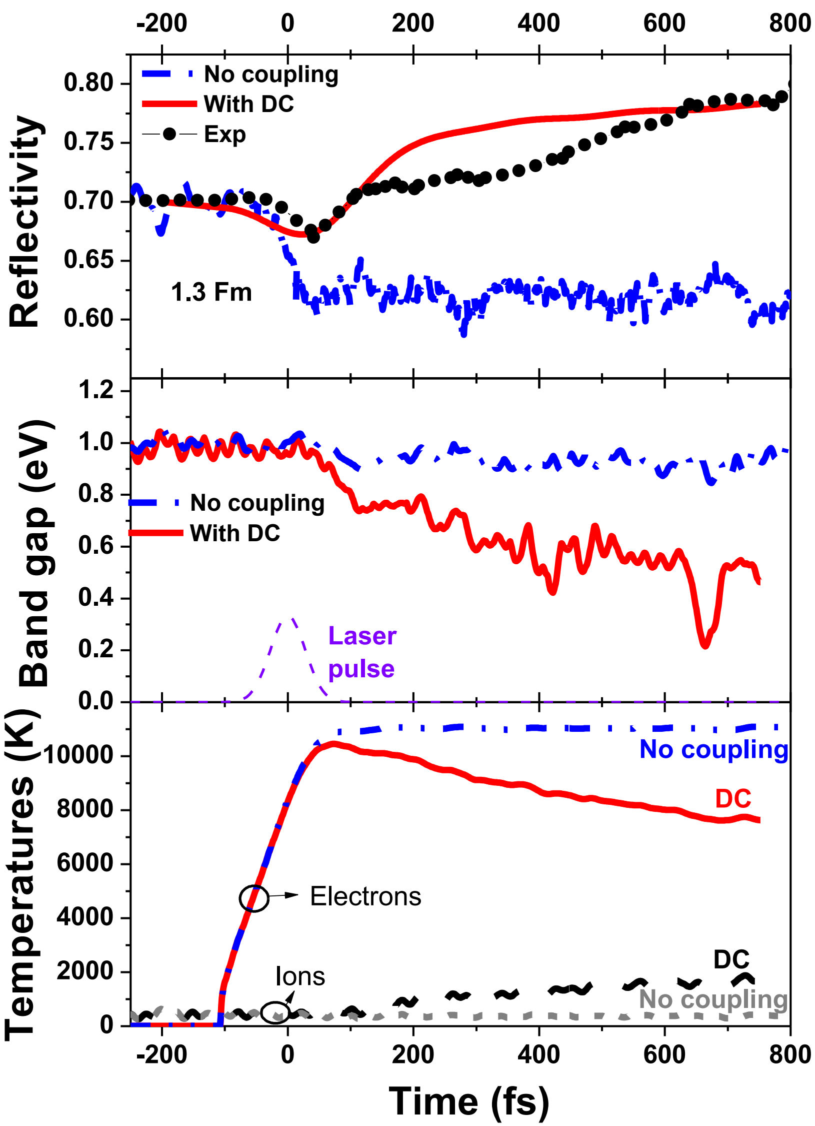


Threshold in terms of incoming photon fluence

corresponding to absorbed dose of 0.6 and 0.9 eV/atom

[N. Medvedev, Z. Li, B. Ziaja, Phys. Rev. B 91 (2015) 054113]

Effects of electron-ion coupling (Si)



Exp: K. Sokolowski-Tinten *et al*,PRB 51(1995) 14186

- Optical properties are affected by shrinking band gap

- Band gap is affected by heating of atoms

- Atoms are heated via electron-ion coupling

Electron-ion coupling is experimentally accessible via pump-probe!

[N. Medvedev *et al.* Phys. Rev. B 95, 014309 (2017)]

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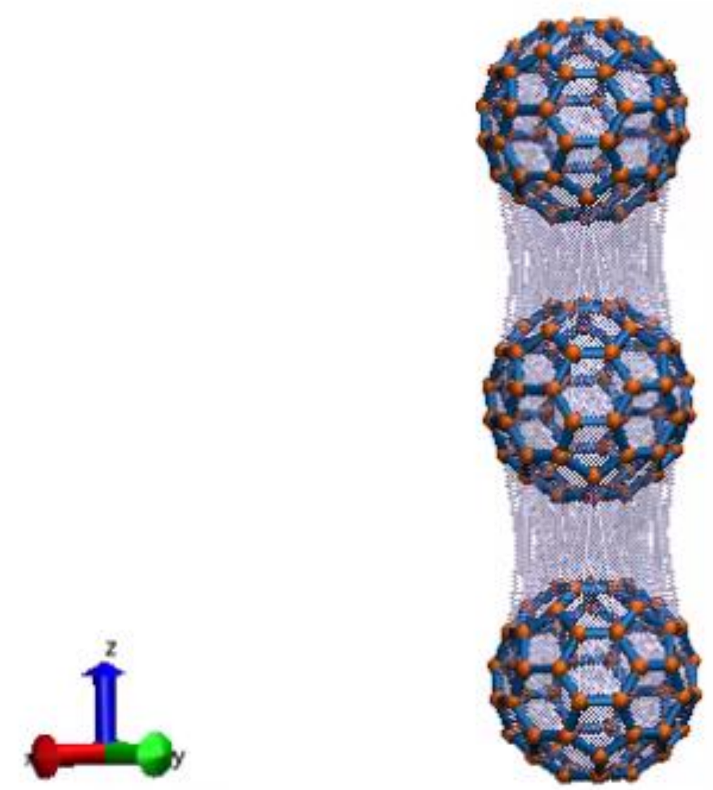
Coulomb explosion of C_{60} crystal

$\hbar\omega$ = 92 keV, FWHM = 30 fs, 0.22 eV/atom



Preliminary results!

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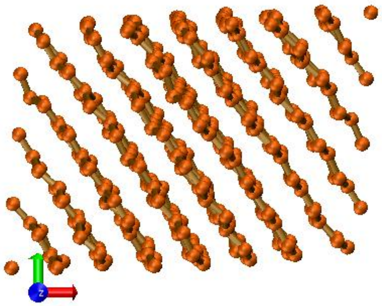
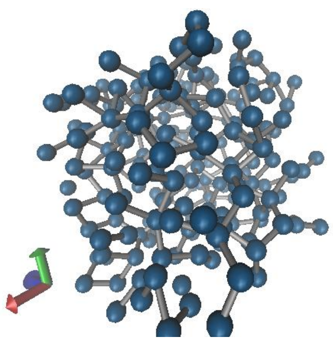
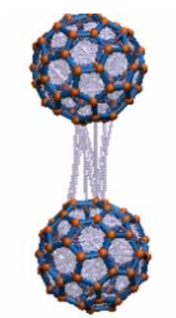
Conclusions



Nonthermal phase transitions: result of change of interatomic potential (~100 fs)



Thermal phase transitions: result of nonadiabatic electron-ion coupling (~1 ps)



Other damage channels also exist, e.g. Coulomb explosion in finite systems

Thank you for you attention!

For details see:

N. Medvedev, H. Jeschke, B. Ziaja, *New J. Phys.* **15**, 015016 (2013)

N. Medvedev, Z. Li, B. Ziaja, *Phys. Rev. B* **91**, 054113 (2015)

N. Medvedev, A. Volkov, B. Ziaja, *NIMB* **365**, 437 (2015)

N. Medvedev, V. Tkachenko, B. Ziaja, *Contrib. Plasma Phys.* **55**, 12 (2015)

N. Medvedev *et al. Phys. Rev.* B **95**, 014309 (2017)