Workshop on transient and ultrafast processes in X-ray excited matter

September 26-27, 2012 DESY, Hamburg

Modeling low photo-excitation of condensed matter

N. Medvedev

CFEL SCIENCE

Center for Free-Electron Laser Science

Outline

1. **Introduction**

2. **Ultrafast electron kinetics:**

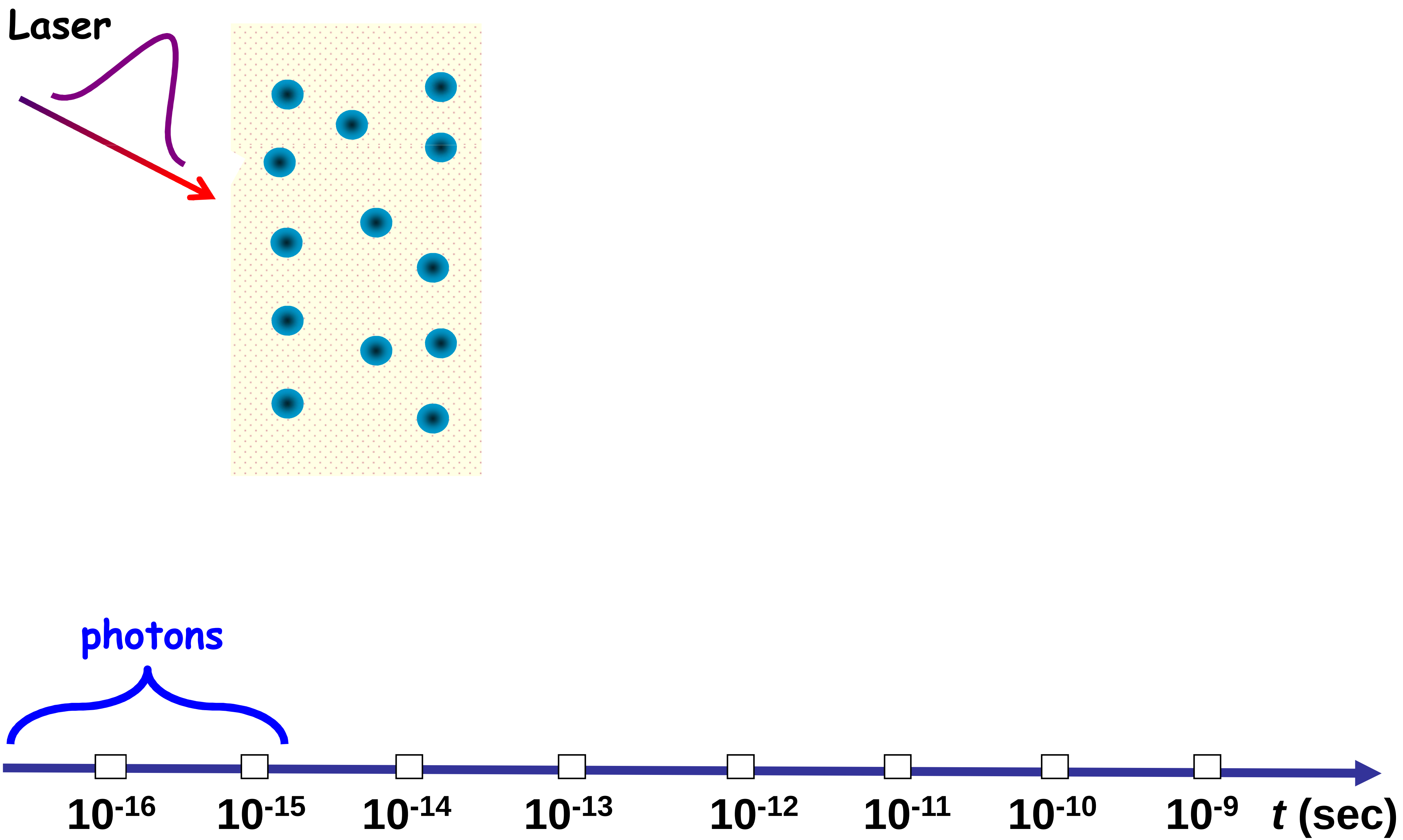
    femtosecond X-ray irradiated SiO2

3. **Ultrafast structural transitions:**

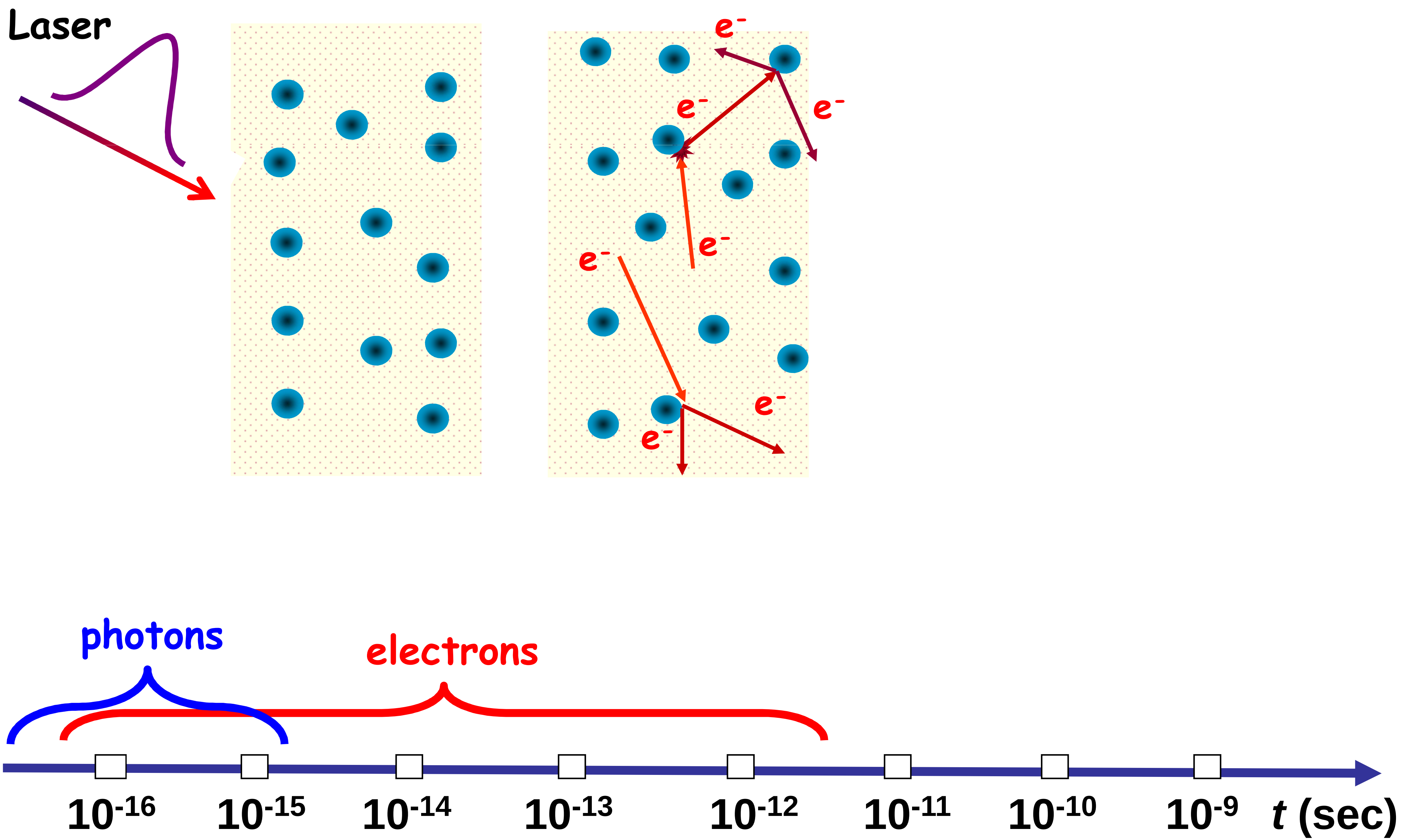
    femtosecond X-ray irradiated Diamond

4. **Summary**

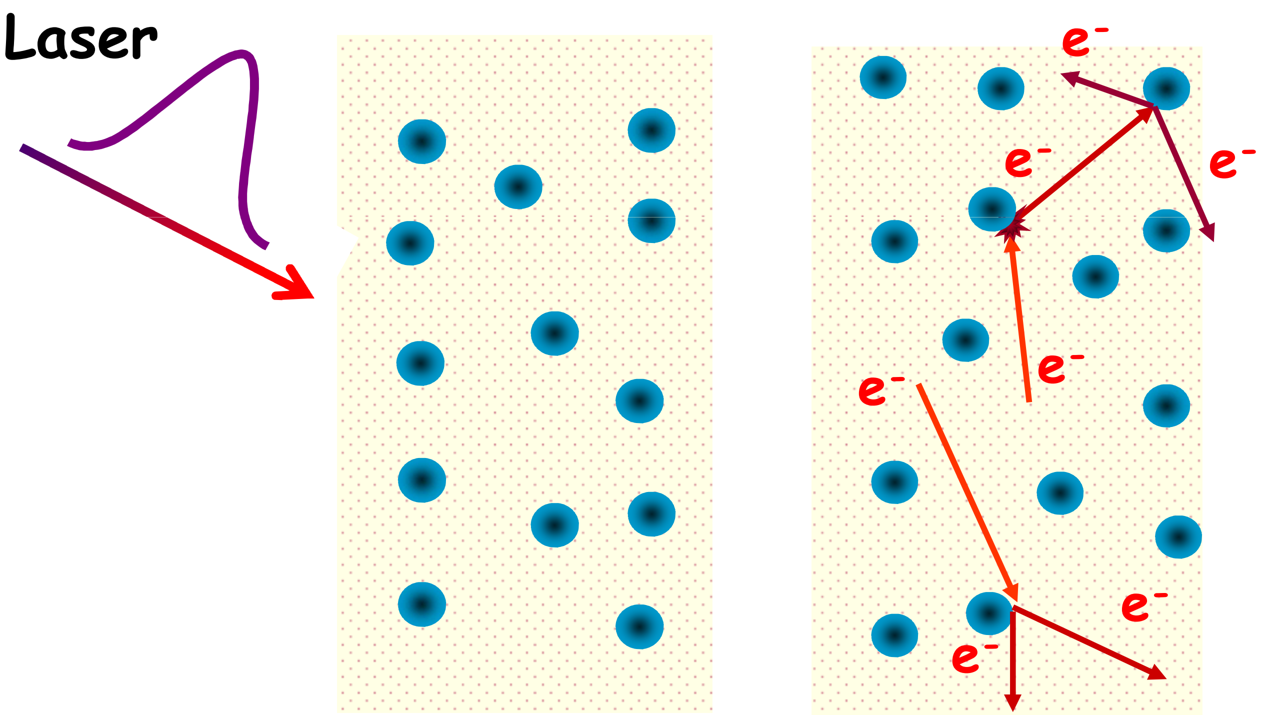
Introduction: timescales

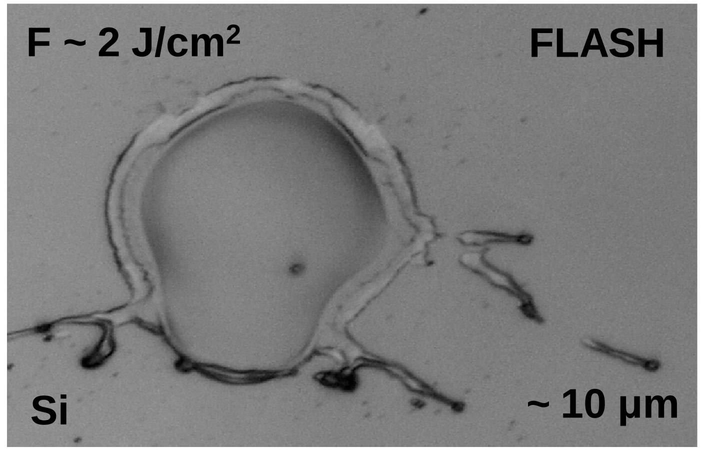


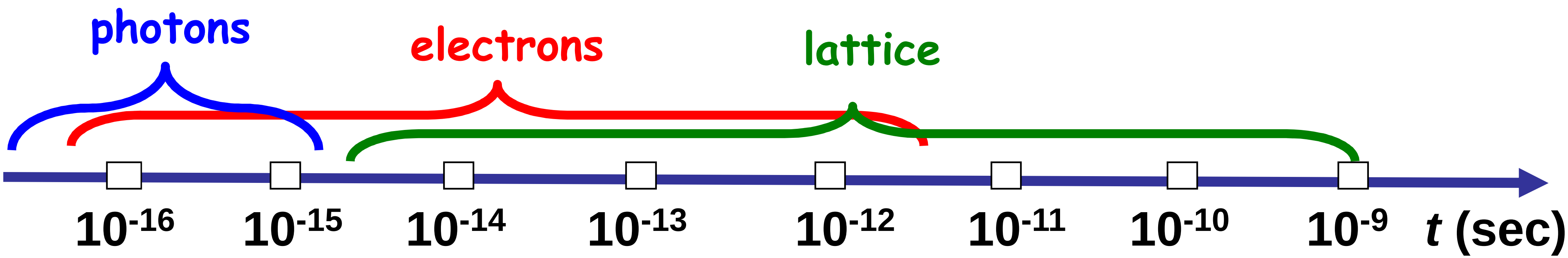
Introduction: timescales



Introduction: timescales

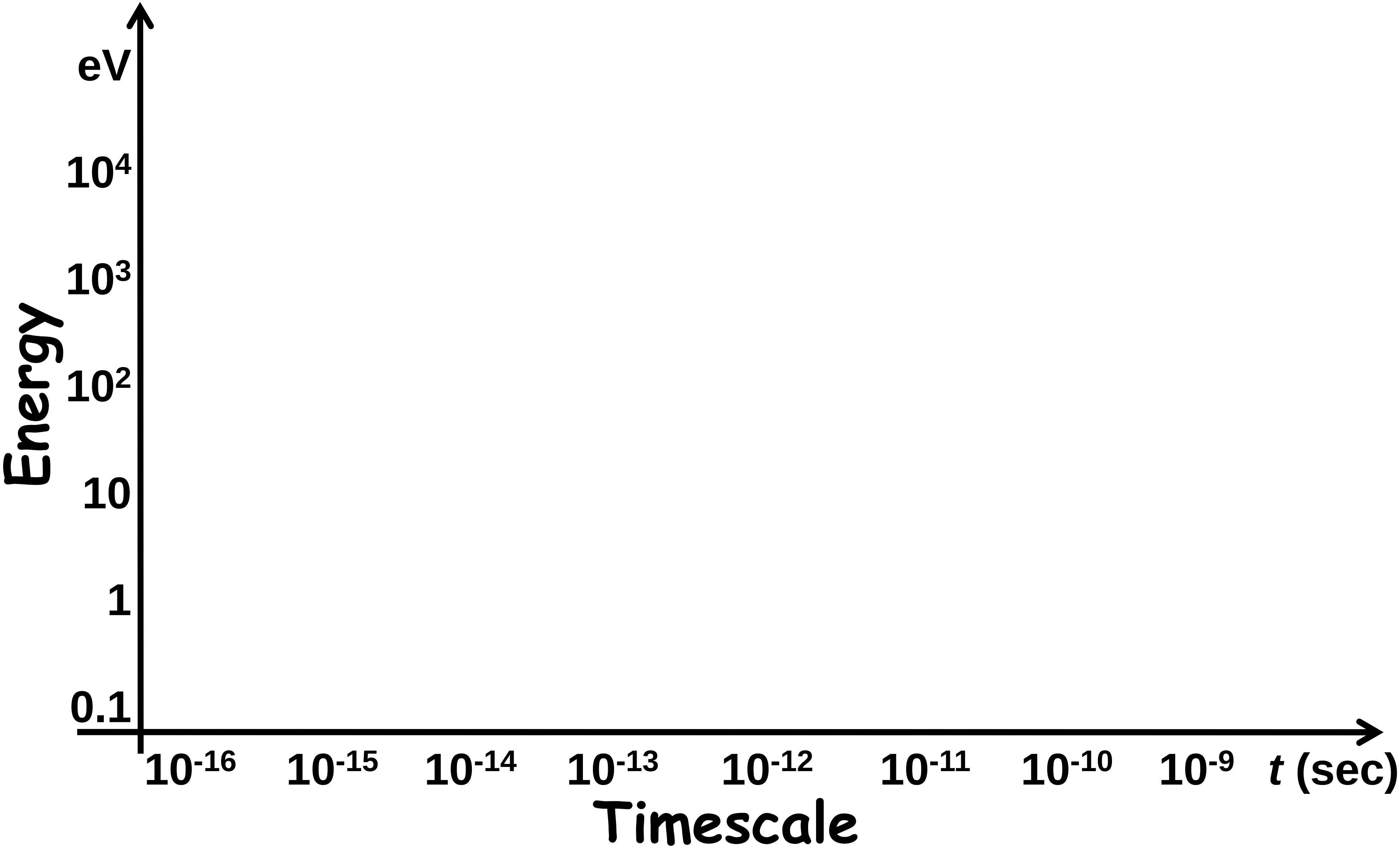




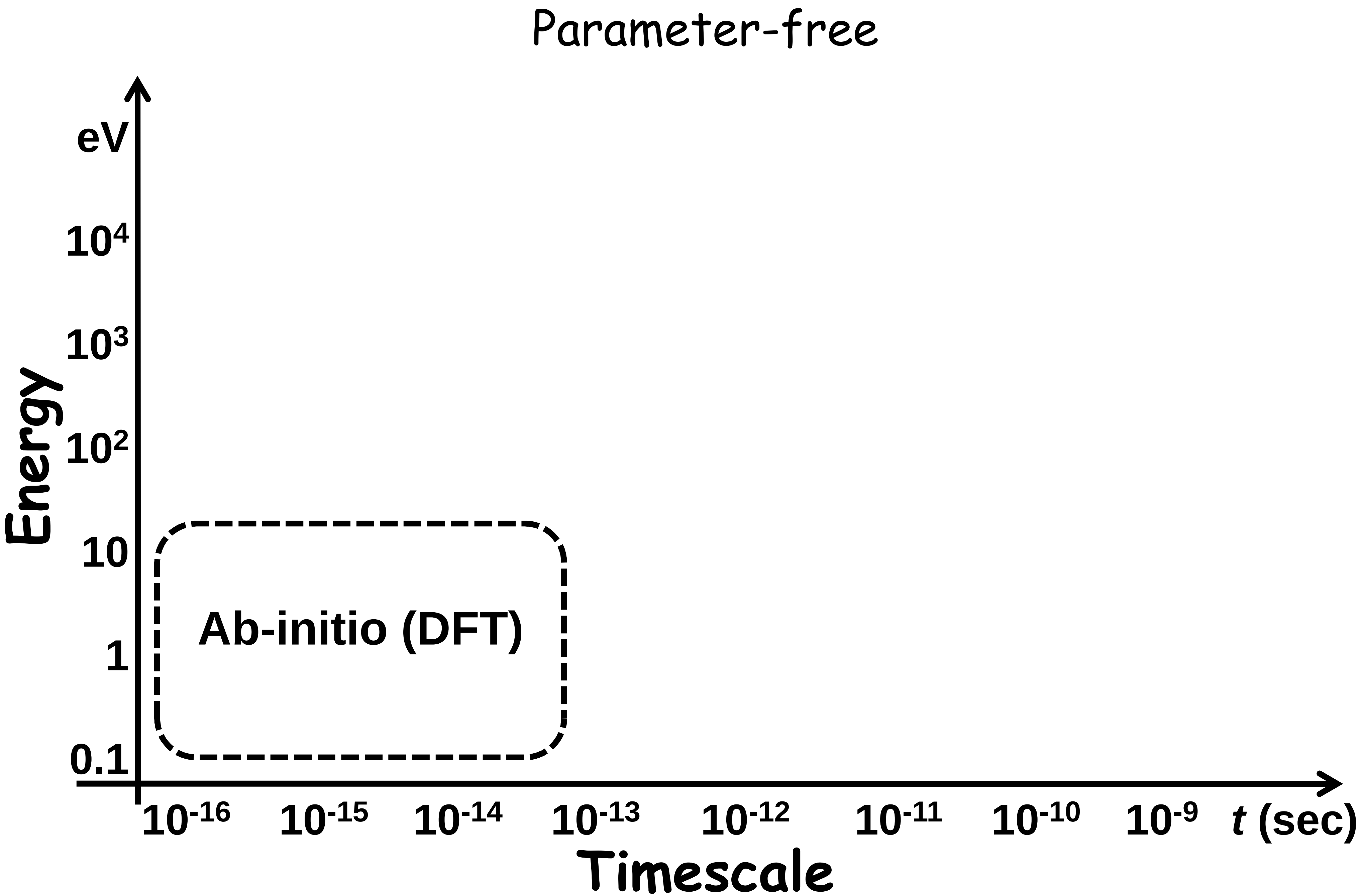


Introduction: common models

Material excitation

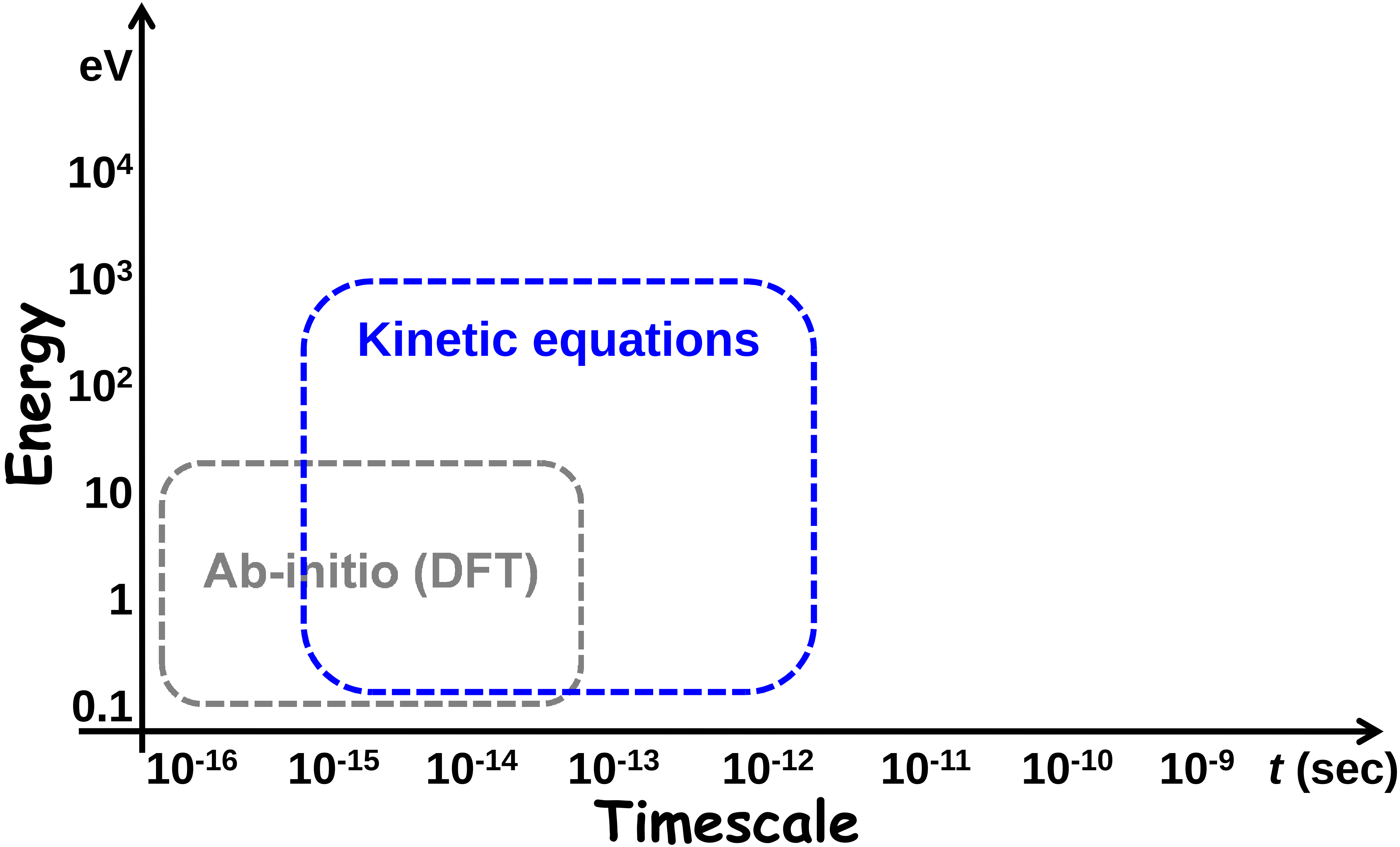


Introduction: common models



Introduction: common models

Needs input: cross-sections/potentials



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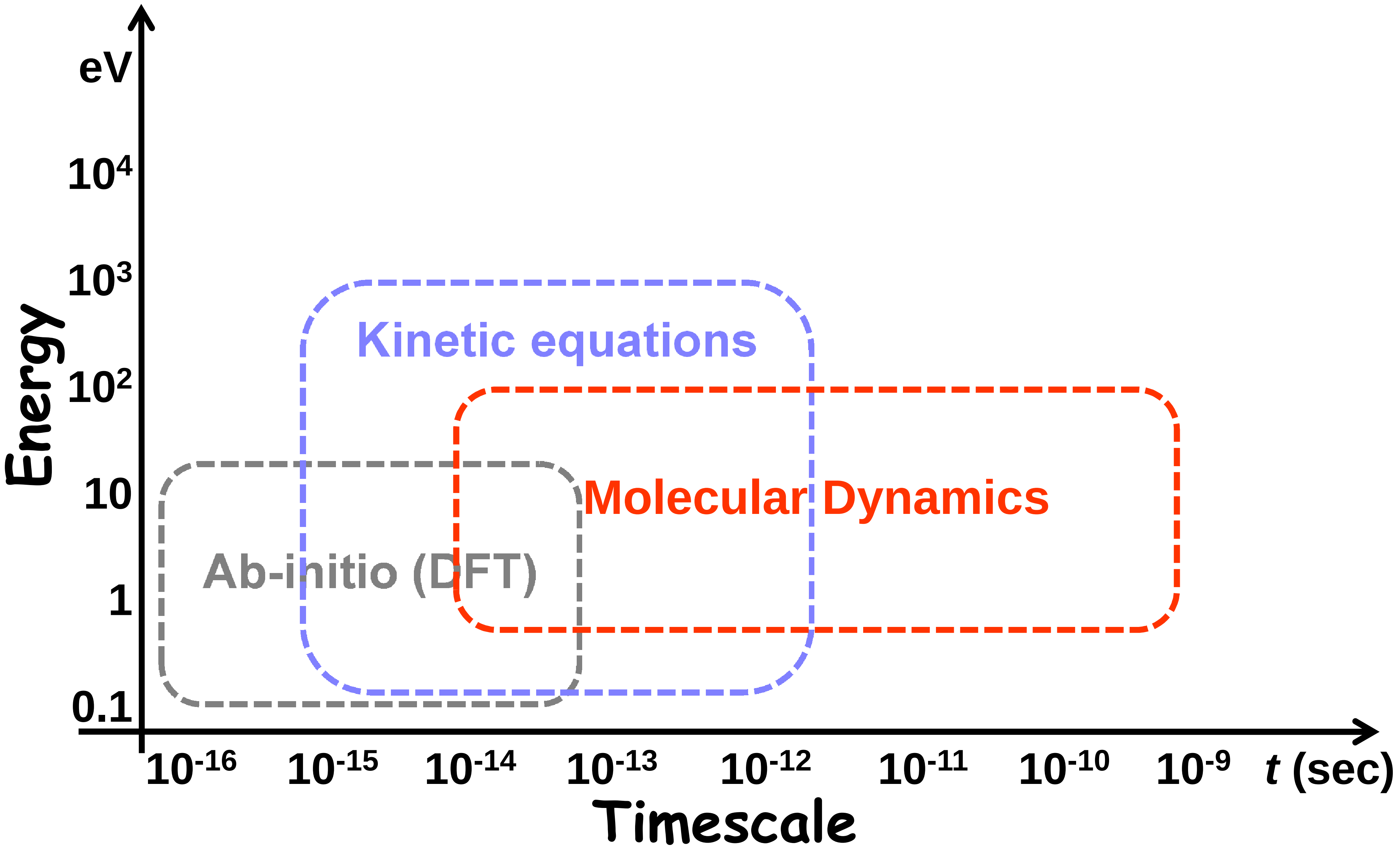
Needs input: potentials



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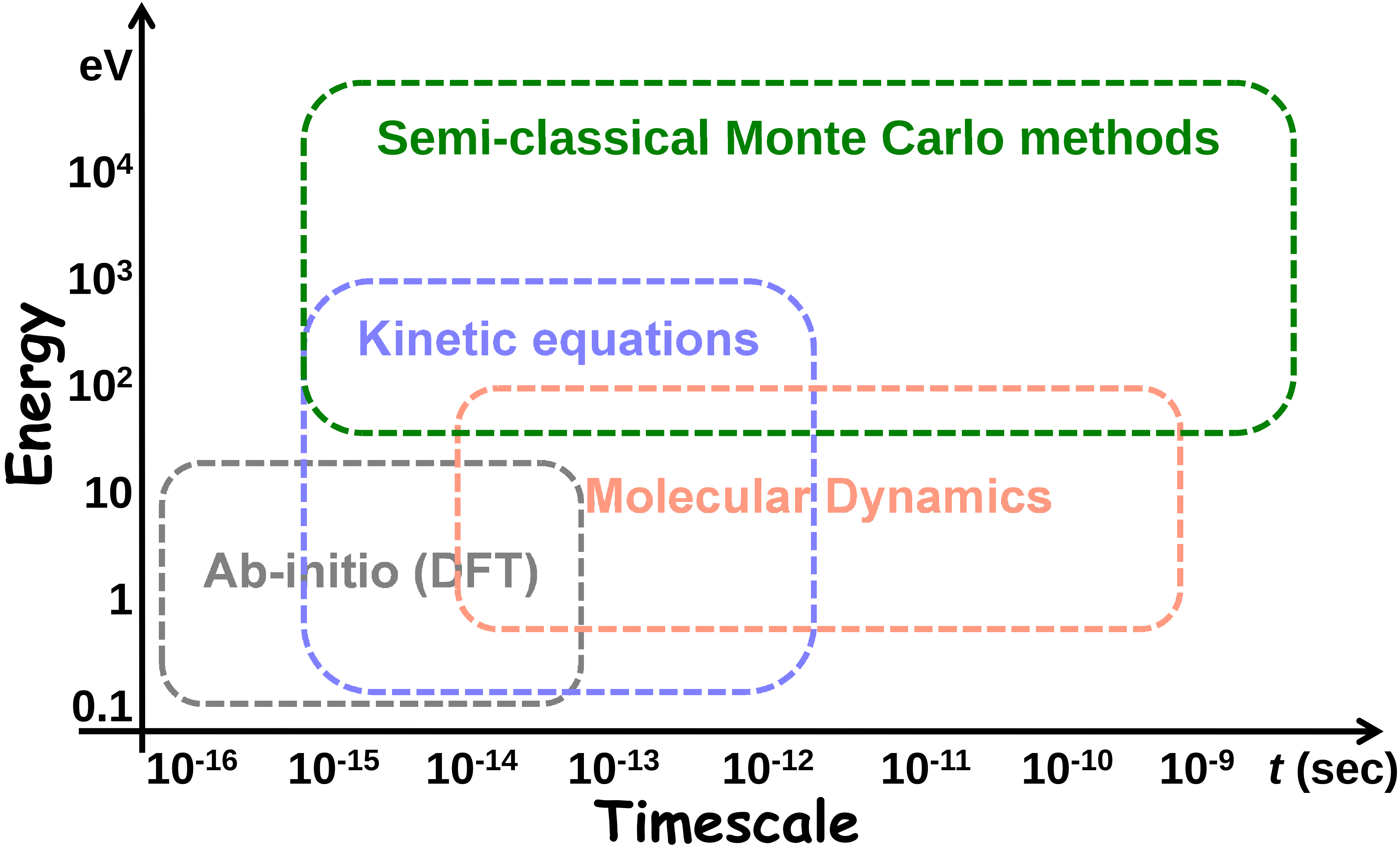
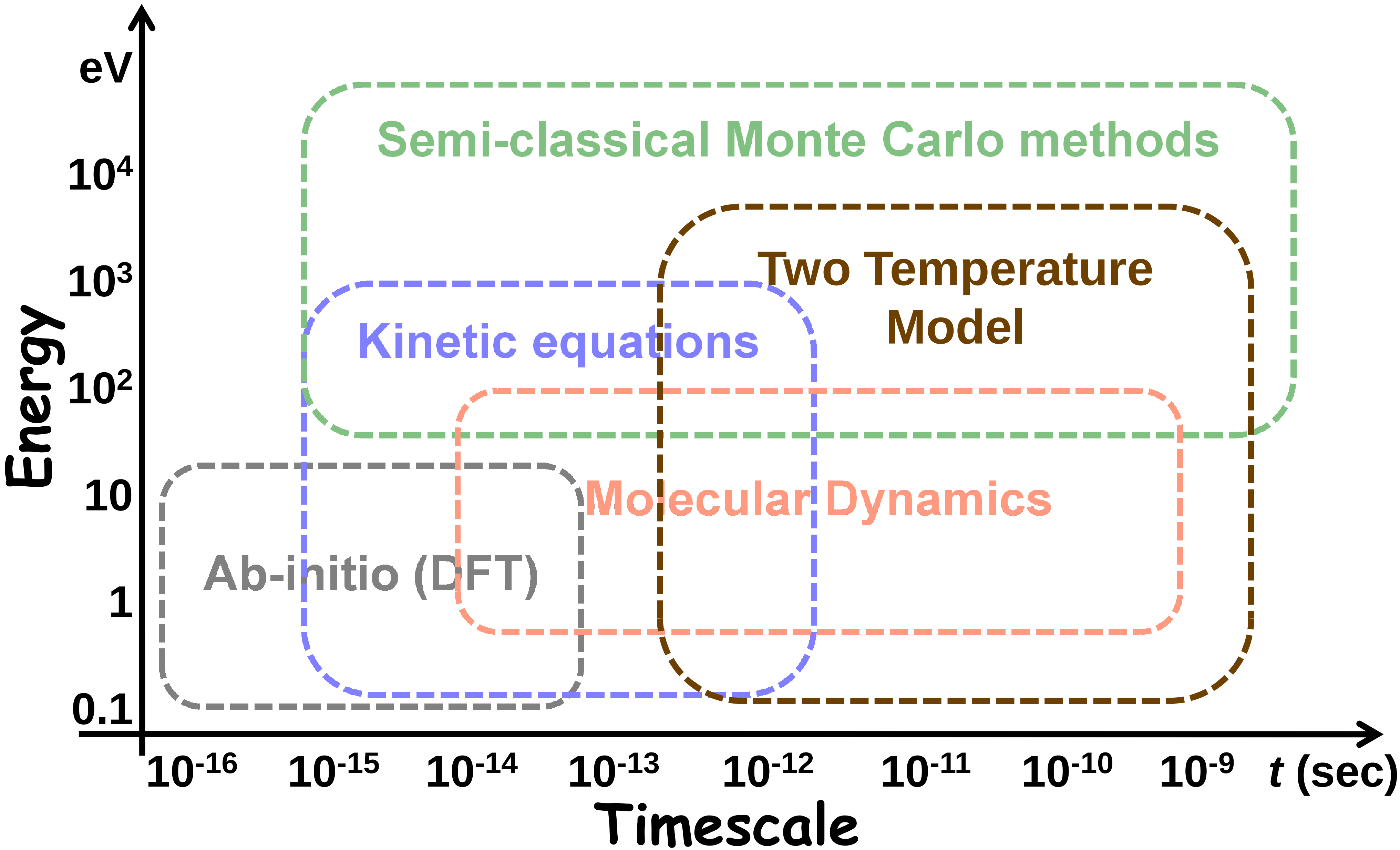


Introduction: common models

Needs input: kinetic coefficients



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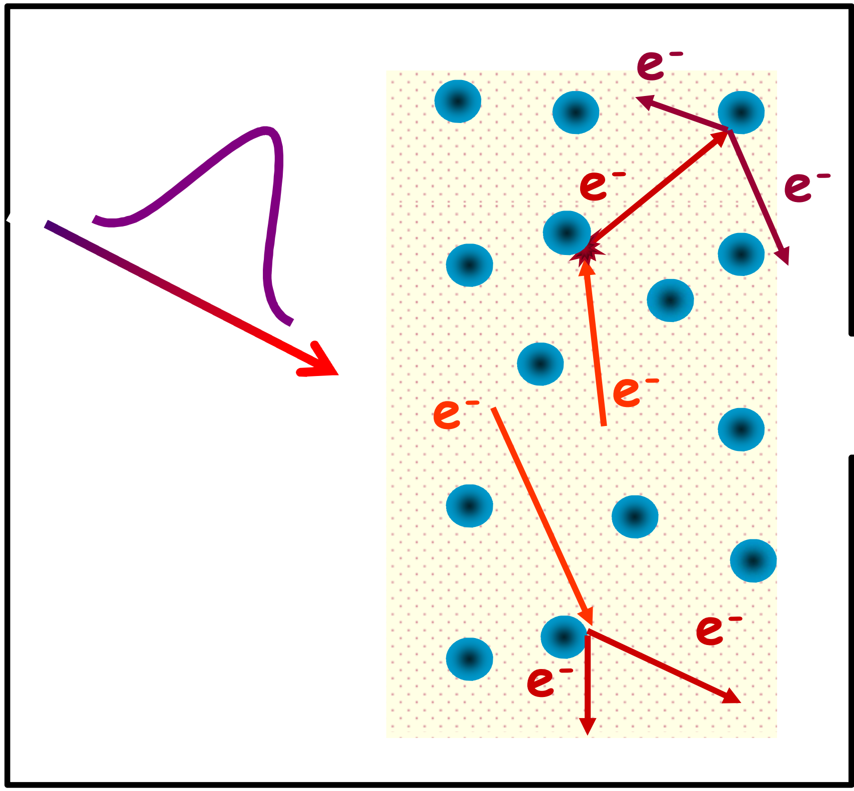
Modeling electron kinetics

MC [N. Medvedev, B. Rethfeld, NJP **12**, 073037 (2010)]



1) electron density

2) CB electron and VB holes distribution functions

Drude model:

$$n(\lambda)^2 = n_0(\lambda)^2 - \left(\frac{\omega_P}{\omega}\right)^2 \frac{1}{1 + i/(\omega\,\tau)}$$

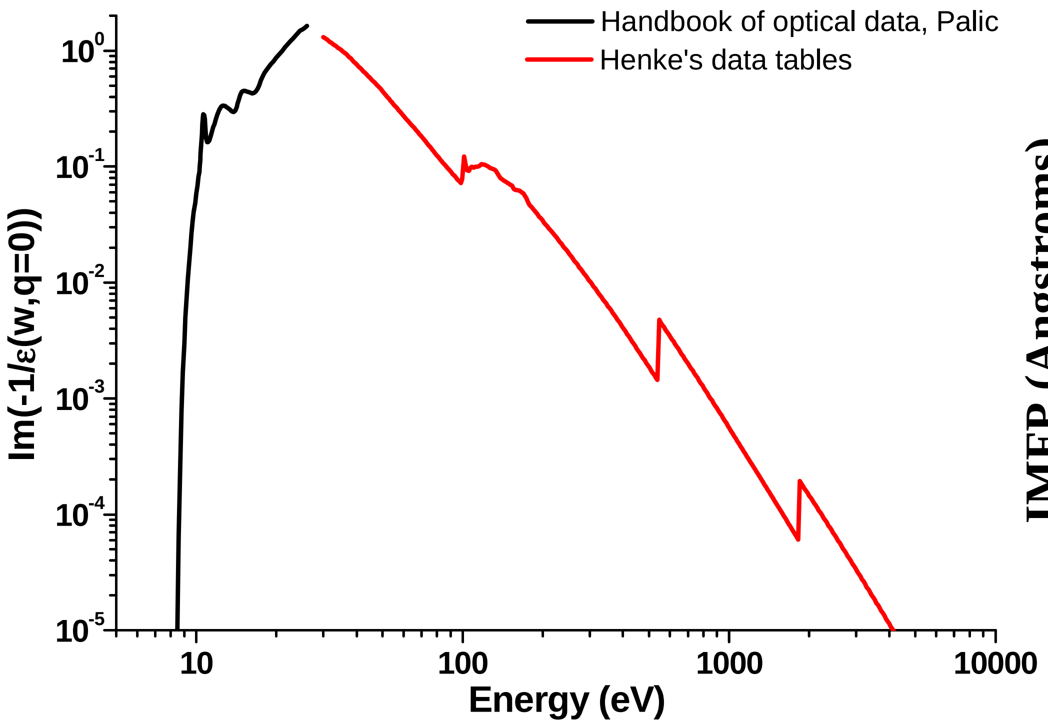
Optical properties: transmission, reflection

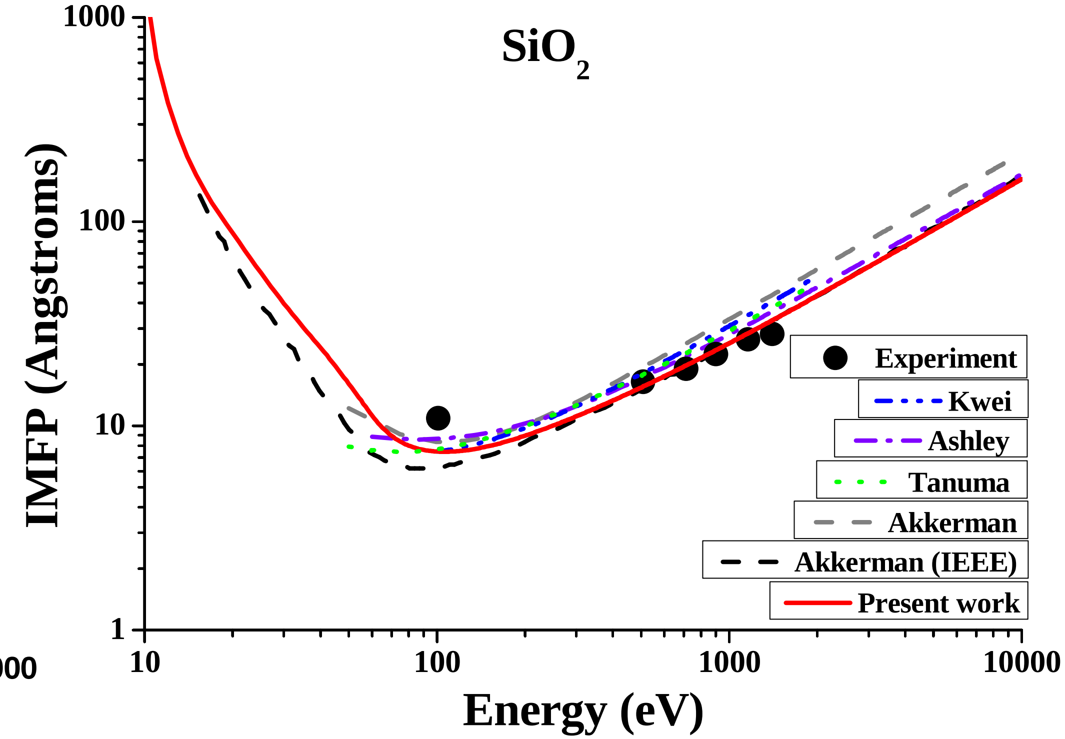
Self-consistent semi-classical Monte-Carlo model

Monte-Carlo cross-sections

Loss-function for *q*=0 => cross-sections for electrons

[R.H. Ritchie and A. Howie, *Philos. Mag.* **36**, 463 (1977)]



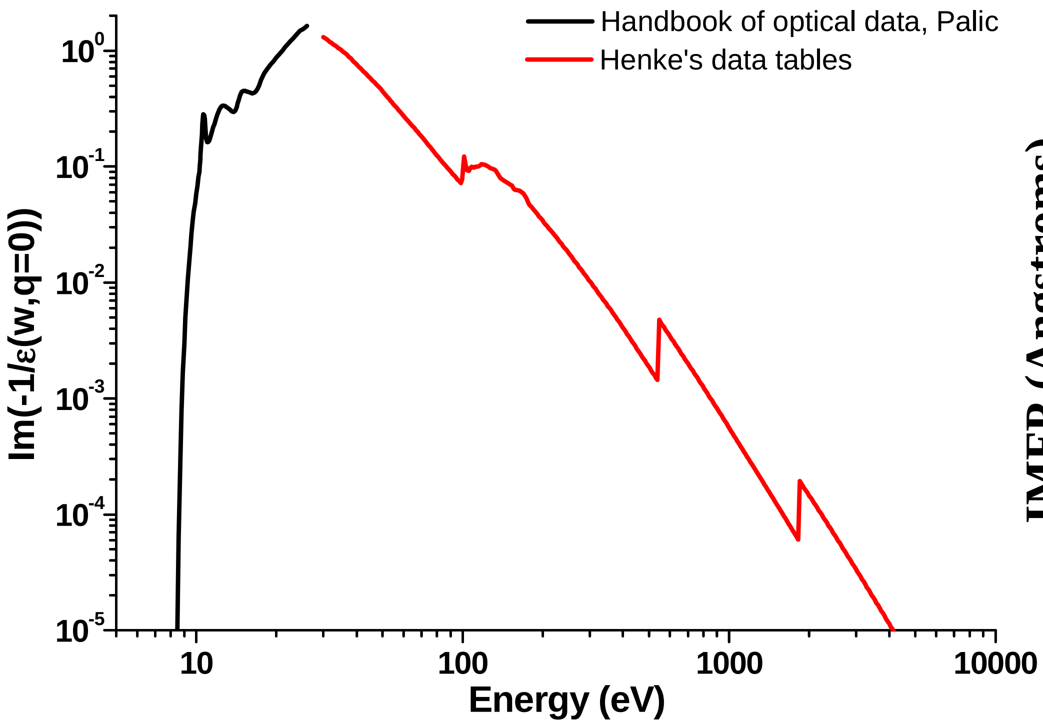


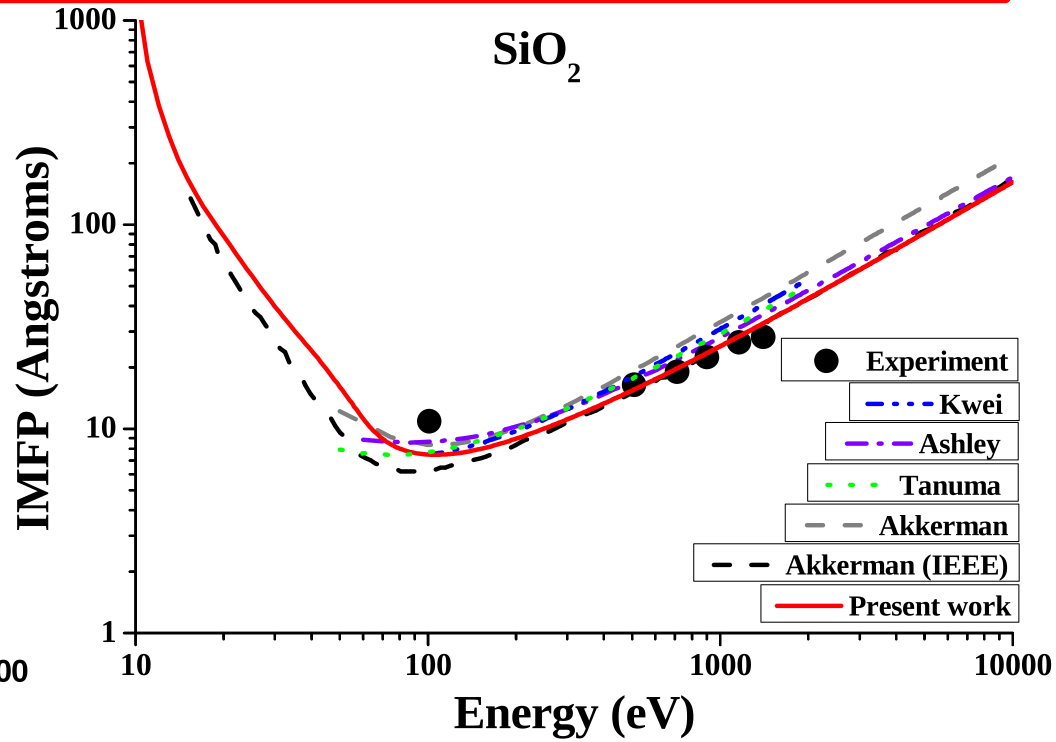
Mean free path used in MC compares well with NIST

[N. Medvedev, *AIP Conf. Proc.* **1464**, 582 (2012)]

Monte-Carlo cross-sections

Highly desirable experiments measuring
optical properties (complex dielectric function / cross-section),
valence and conduction band structure / DOS
as functions of photon energy and fluence





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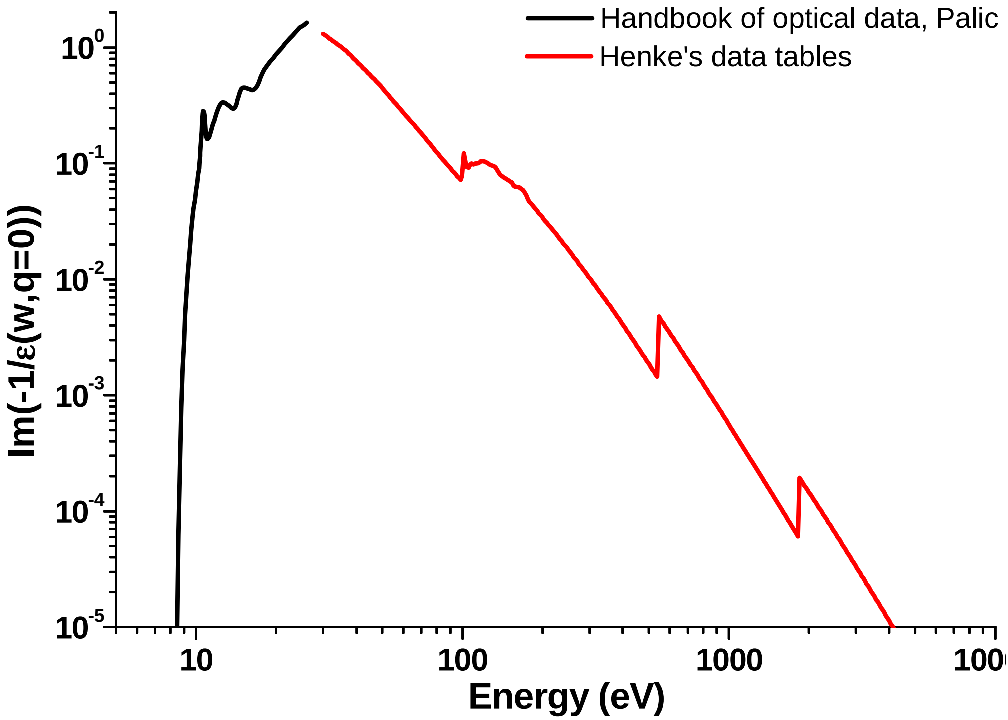

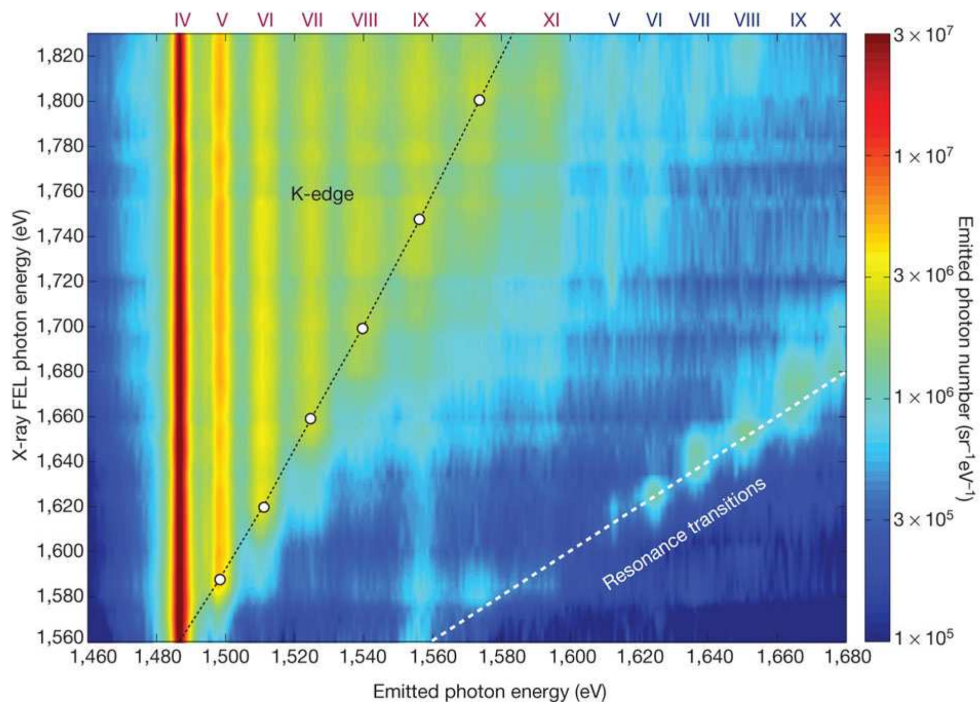

S.M. Vinko *et al. Nature* **482**, 59 (2012)

DOS: S.M. Vinko *et al. PRL* **104**, 225001 (2010)

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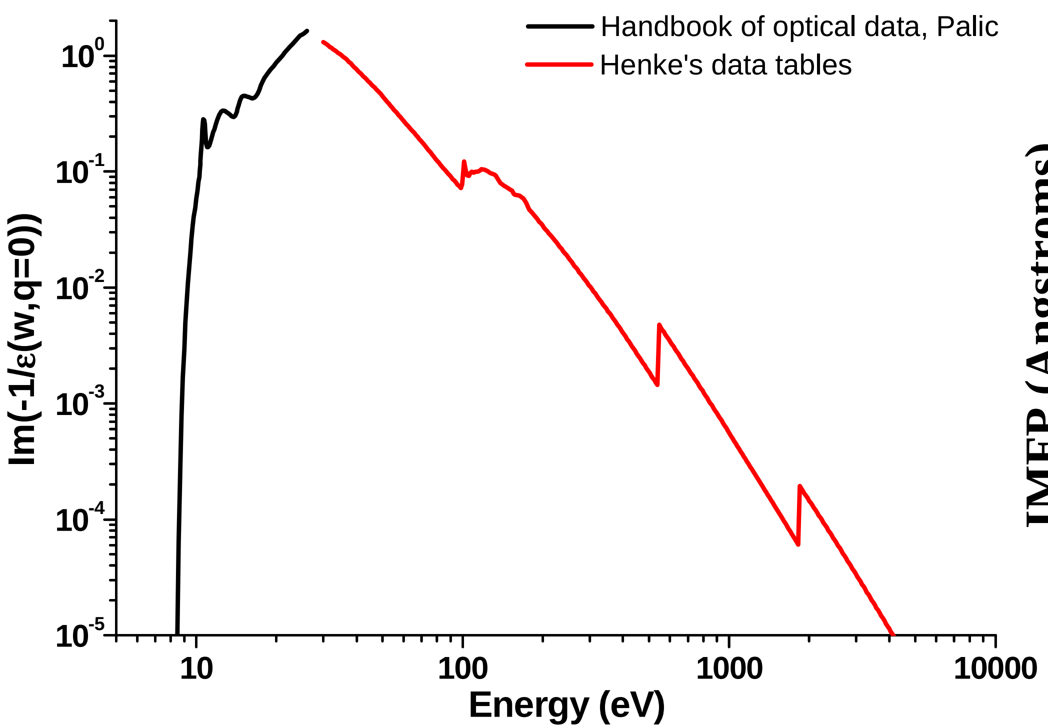
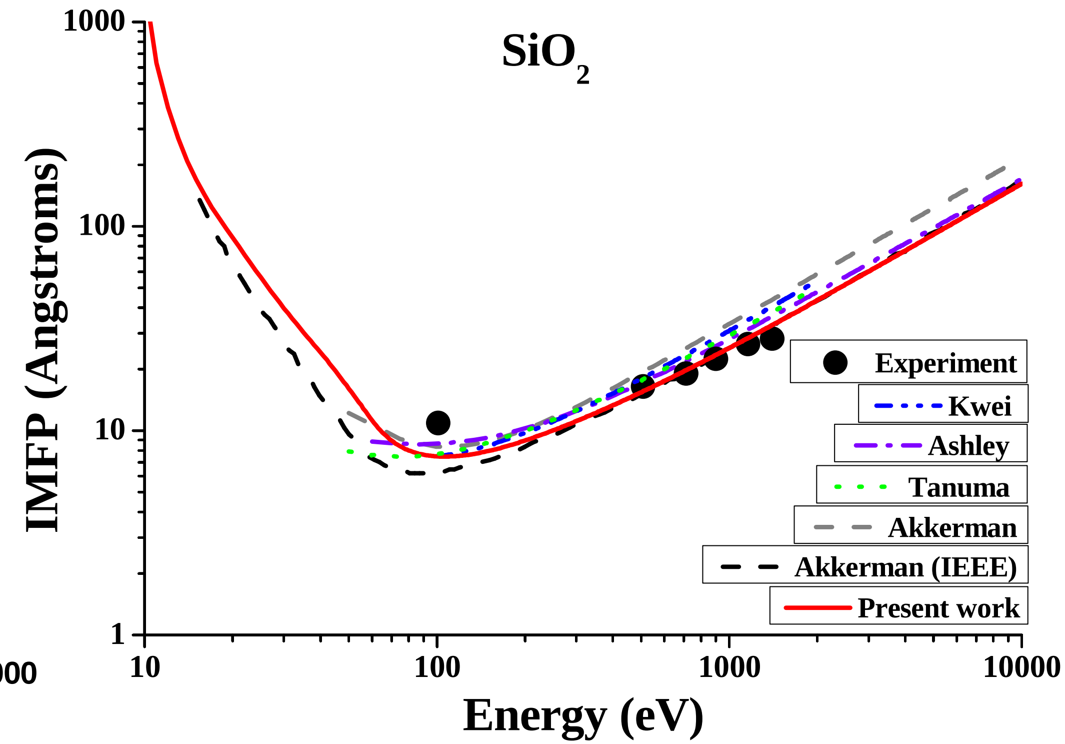
Loss-function for *q*=0 => cross-sections for electrons

[R.H. Ritchie and A. Howie, *Philos. Mag.* **36**, 463 (1977)]

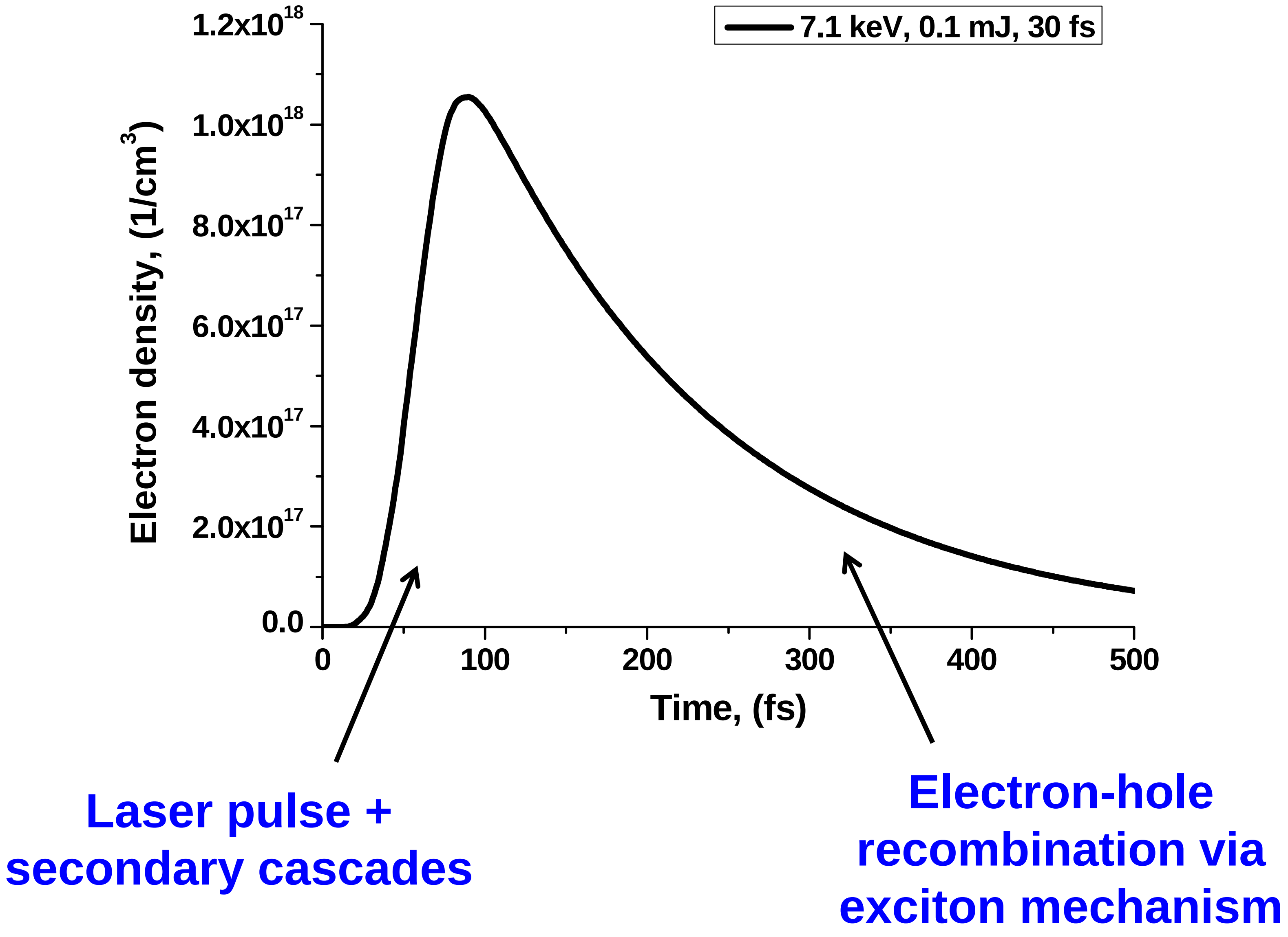




Mean free path used in MC compares well with NIST

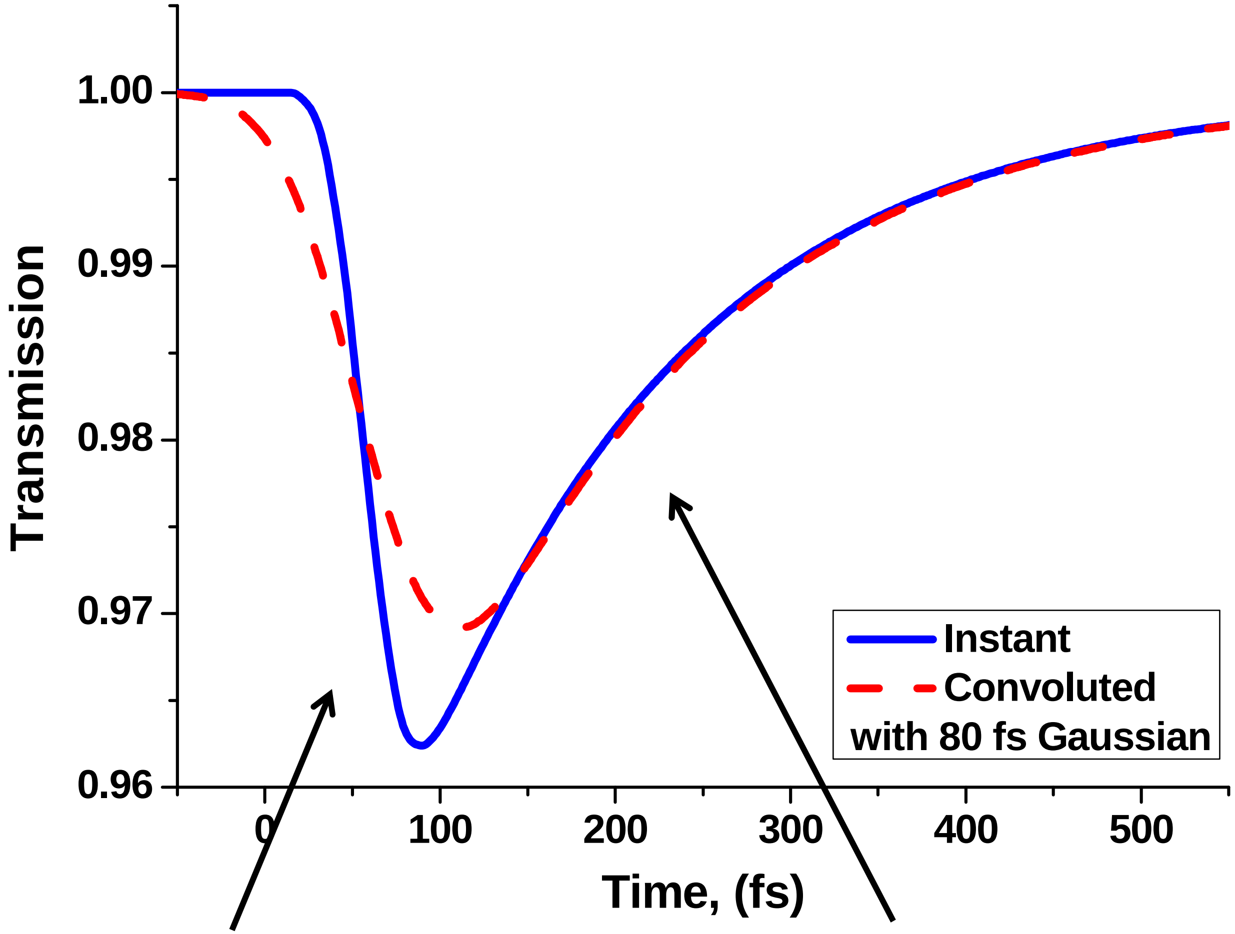
[N. Medvedev, *AIP Conf. Proc.* **1464**, 582 (2012)]

Results: electron density



N. Medvedev, B. Ziaja, *et. al*. Contr. to Plasma Phys. (*submitted*)

Results: transmission



Laser pulse + secondary cascades

Electron-hole recombination via exciton mechanism

N. Medvedev, B. Ziaja, *et. al*. Contr. to Plasma Phys. (*submitted*)

Results: comparison of transmission

Experiments are needed measuring not only electron density,

[C. Gahl *et al.*, *Nature Photonics* **2**, 165 (2008)]

but also

- **Temperature**
- **Distribution function**

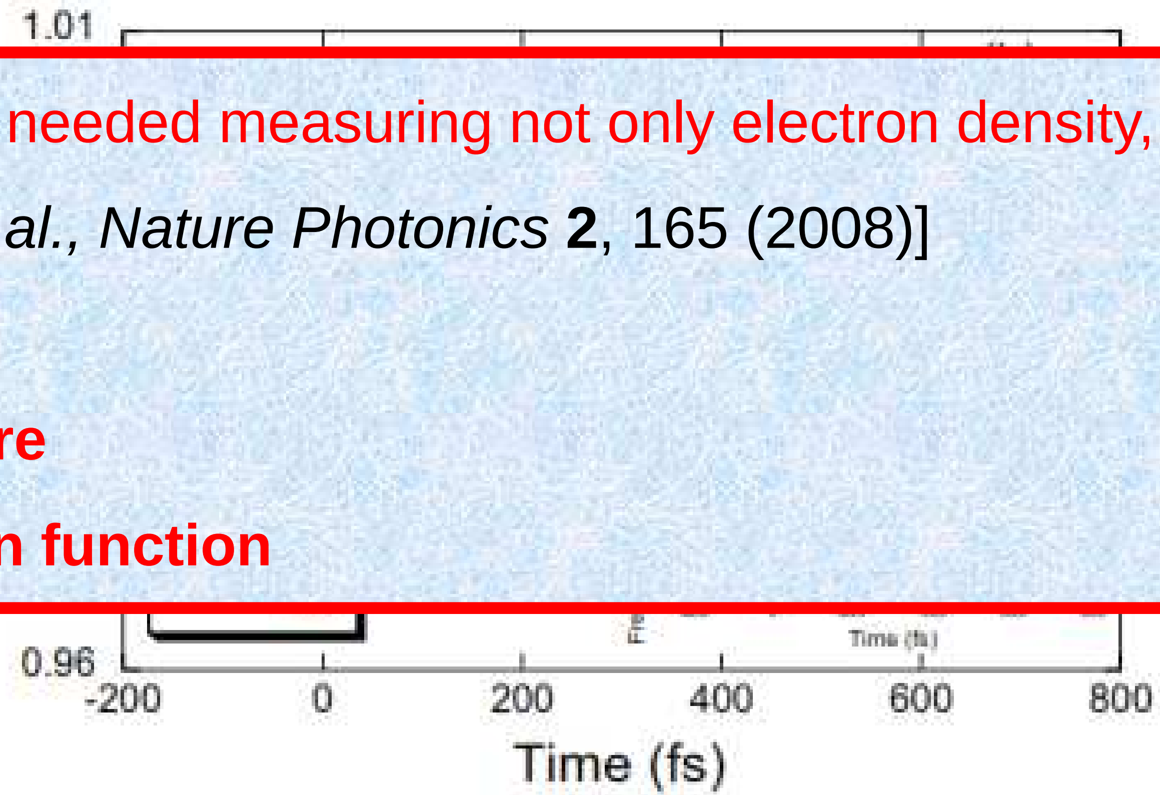


Figure 3: (a) Monte-Carlo calculation of the free carrier density obtained on SiO_2 glass slab with a 7.1 keV unfocused 30 fs FEL beam. (b) Calculation of the transmission by using the Drude model coupled with MC density (figure (a)) and compared with a single-shot experimental data on glass.

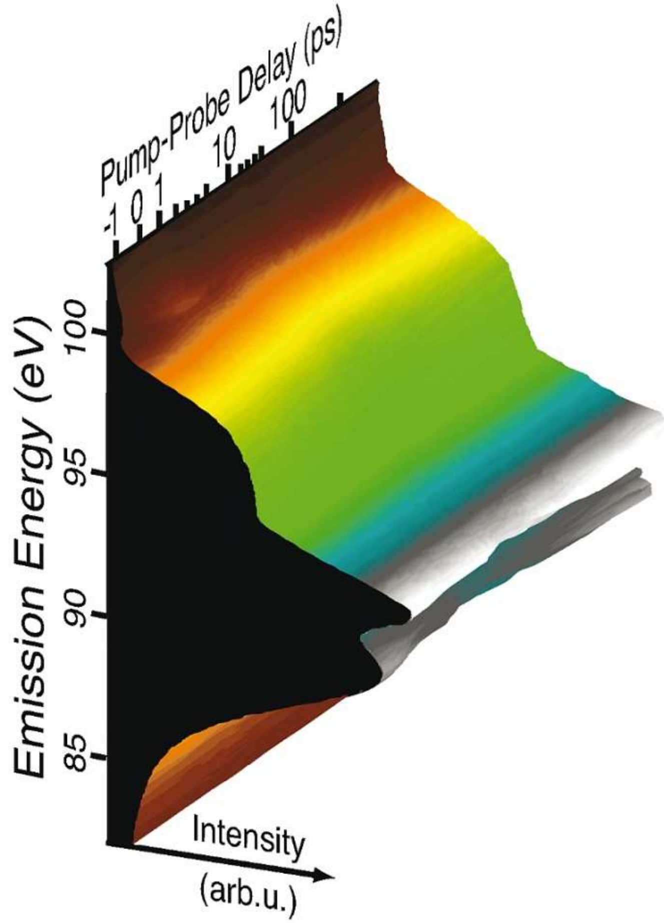
Results: comparison of transmission

Experiments are needed measuring not only electron density, but also

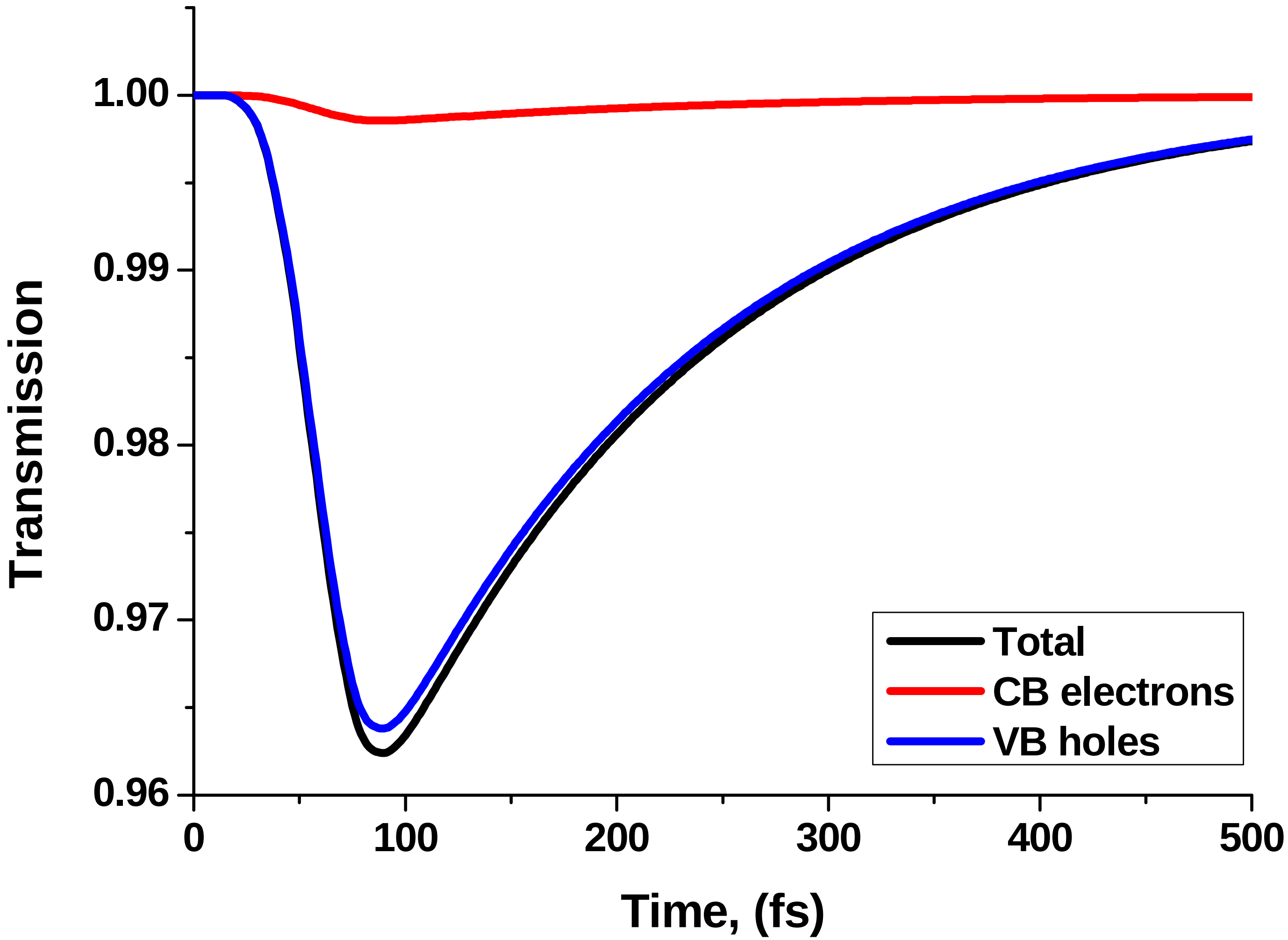
- **Temperature**
- **Distribution function**

Snapshots of valence electrons in Si irradiated with optical pulse , probed with FLASH

M. Beye *et al.*, *Proc. Natl. Acad. Sci. USA* **107**, 39 (2010)



Results: what do we learn?

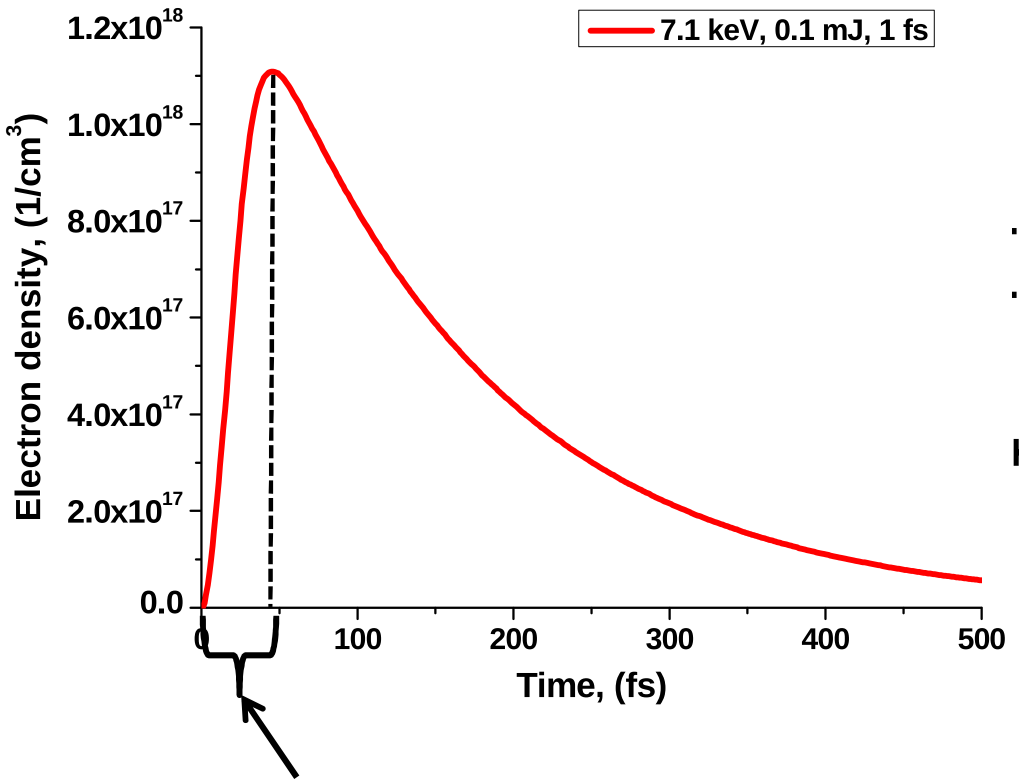


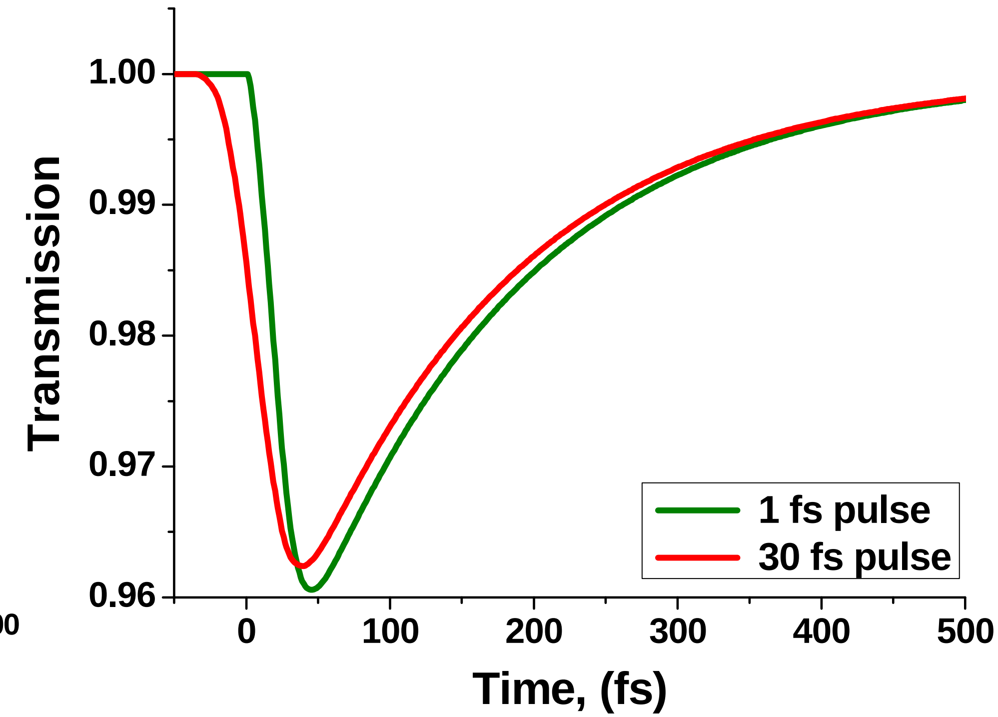
Electrons are too fast, contribution of valence holes is dominant

N. Medvedev, B. Ziaja, *et. al*. Contr. to Plasma Phys. (*submitted*)

Results: what do we learn?

Making pulse shorter: 1 fs





Cascades of secondary electrons – a physical limitation on temporal resolution

The higher photon energy is, the longer it takes for cascading!

N. Medvedev, B. Ziaja, *et. al*. Contr. to Plasma Phys. (*submitted*)

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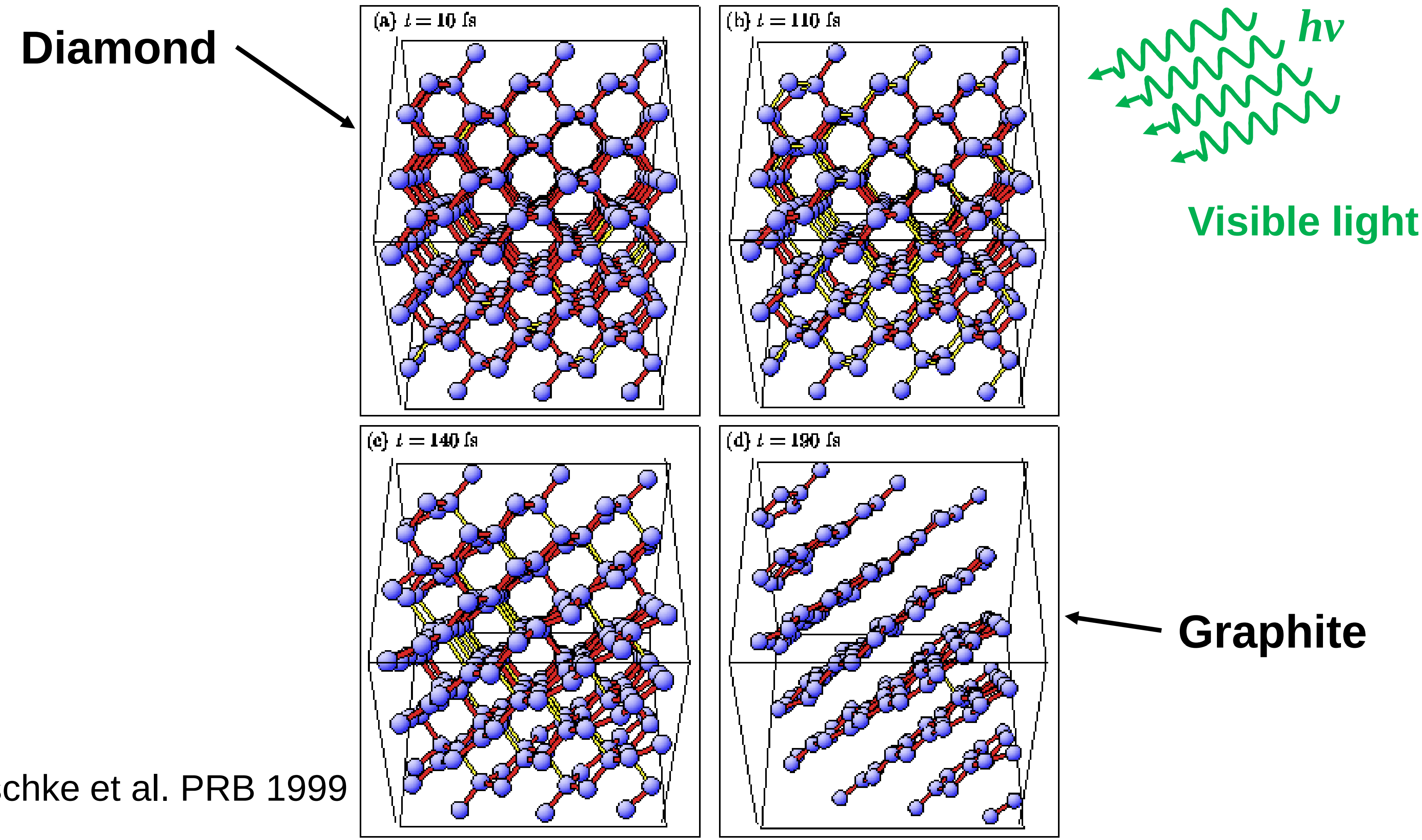
Nonthermal melting of semiconductors

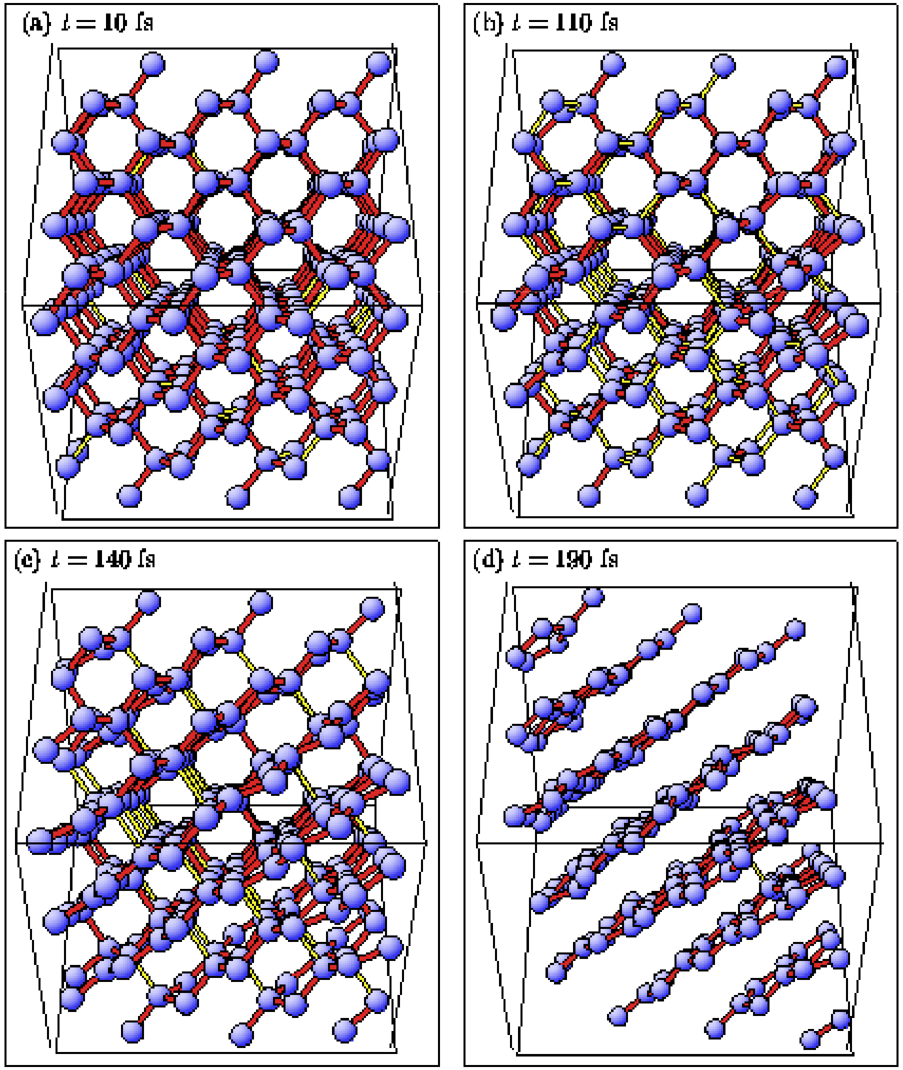


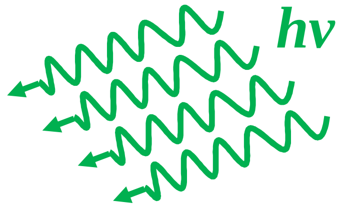
H. Jeschke et al. PRB 1999

Ultrafast phase transition due to a change of interatomic potential

Nonthermal melting of semiconductors

Diamond





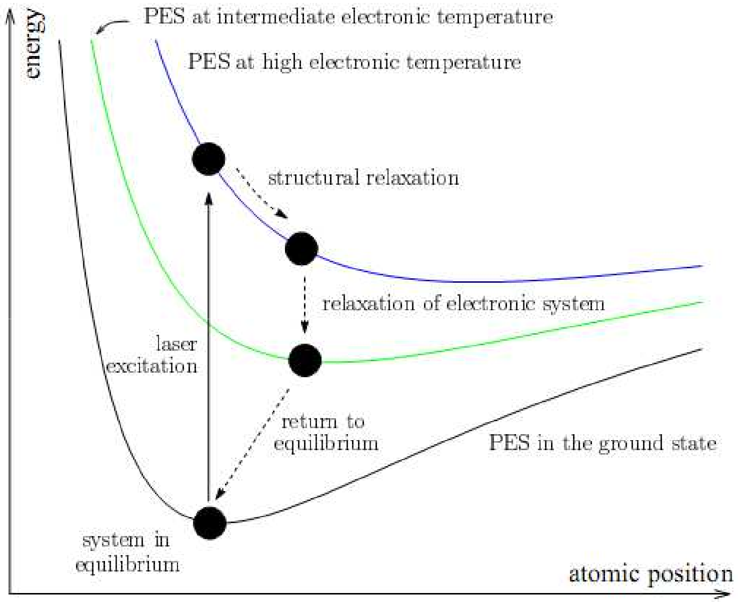
Visible light

Graphite

H. Jeschke et al. PRB 1999

Experimentally: A. Rousse *et al.*, *Nature* **410**, 65 (2001)

Tight-binding Molecular dynamics (TBMD)



$$m_k \ddot{\mathbf{r}}_k = -\frac{\partial \Phi(\{r_{ij}\}, t)}{\partial \mathbf{r}_k}$$

Electrons — **Core repulsion**

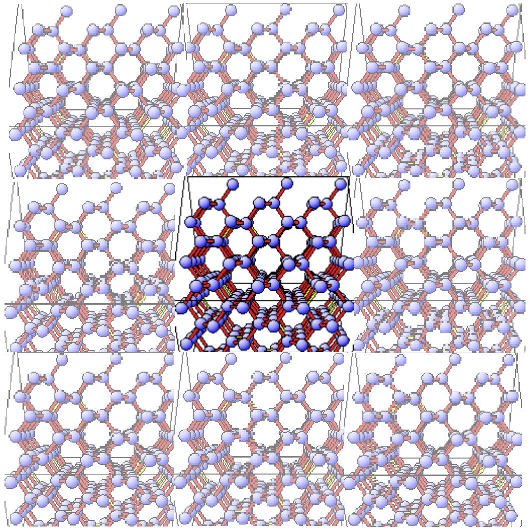
$$\Phi(\{r_{ij}(t)\}, t) = \sum_m f(\epsilon_m, t)\epsilon_m + \frac{1}{2}\sum_{\substack{ij \\ j \neq i}} E_{\text{rep}}(r_{ij})$$

$f(\epsilon_m, t)$ - transient electron distribution function

$\epsilon_m(\{r_{ij}(t)\}) = \langle m | H_{\text{TB}}(\{r_{ij}(t)\}) | m \rangle$ **- transient band structure**

[H. Jeschke et al. PRB 1999]

Parrinello-Rahman molecular dynamics



Periodic boundaries

$$L = \sum_{i=1}^{N} \frac{m_i}{2} \dot{\mathbf{s}}_i^{\mathrm{T}} h^{\mathrm{T}} h \, \dot{\mathbf{s}}_i + K_{\mathrm{cell}} - \Phi(\{r_{ij}\}, t) - U_{\mathrm{cell}}$$

$$\ddot{\mathbf{s}}_i = -\frac{1}{m_i} \sum_{j \neq i} \frac{\partial \Phi(r_{ij})}{\partial r_{ij}} \frac{\mathbf{s}_i - \mathbf{s}_j}{r_{ij}} - g^{-1} \dot{g} \dot{\mathbf{s}}_i$$

Changing super-cell size and shape

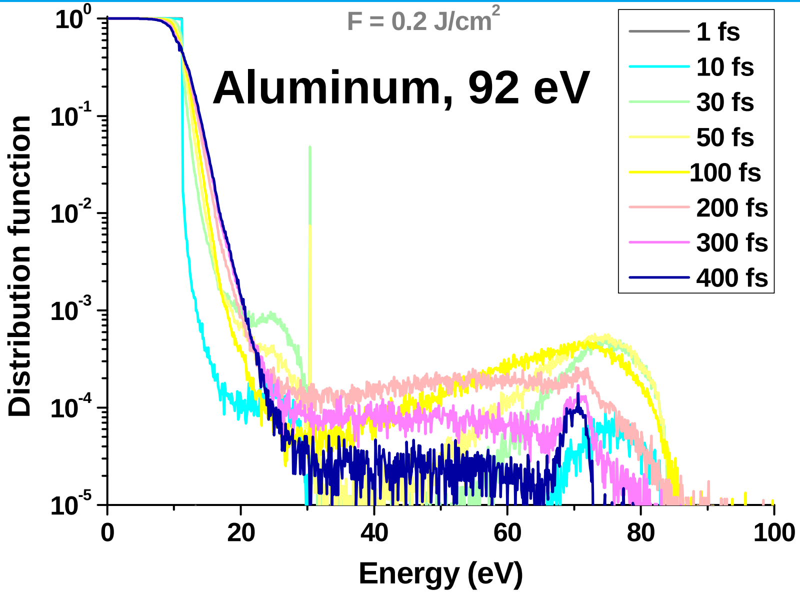
$$\Phi(\{r_{ij}(t)\}, t) = \sum_m f(\epsilon_m, t) \epsilon_m + \frac{1}{2} \sum_{\substack{ij \\ j \neq i}} E_{\mathrm{rep}}(r_{ij})$$

$f(\epsilon_m, t)$ - transient electron distribution function

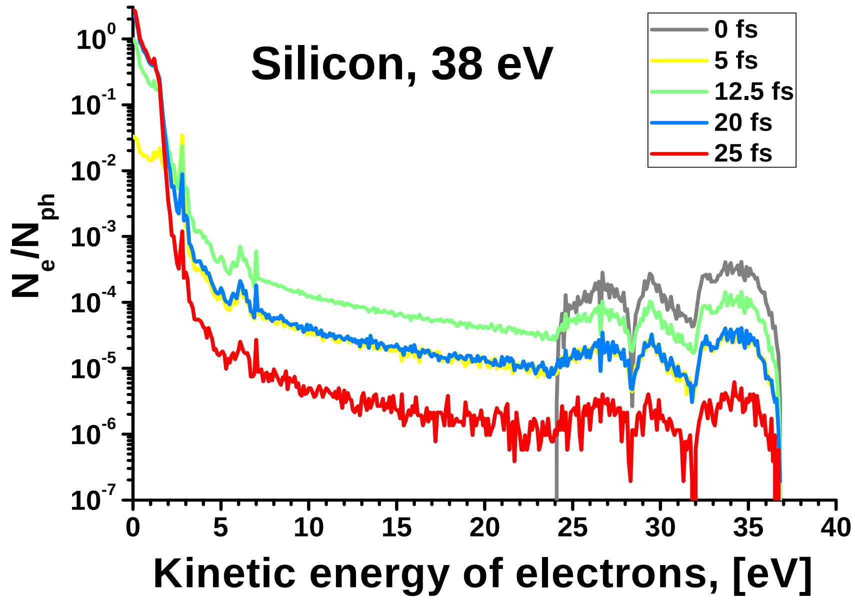
$\epsilon_m(\{r_{ij}(t)\}) = \langle m | H_{\mathrm{TB}}(\{r_{ij}(t)\}) | m \rangle$ **- transient band structure**

[M. Parrinello and A. Rahman, PRL 1980]

Electron distribution function

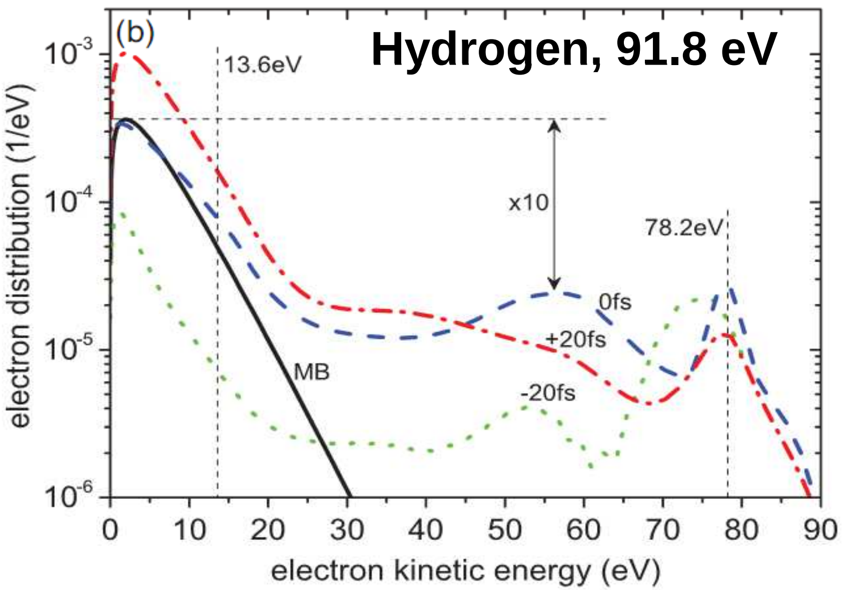


[N. Medvedev *et al.*, PRL **107** (2011)]

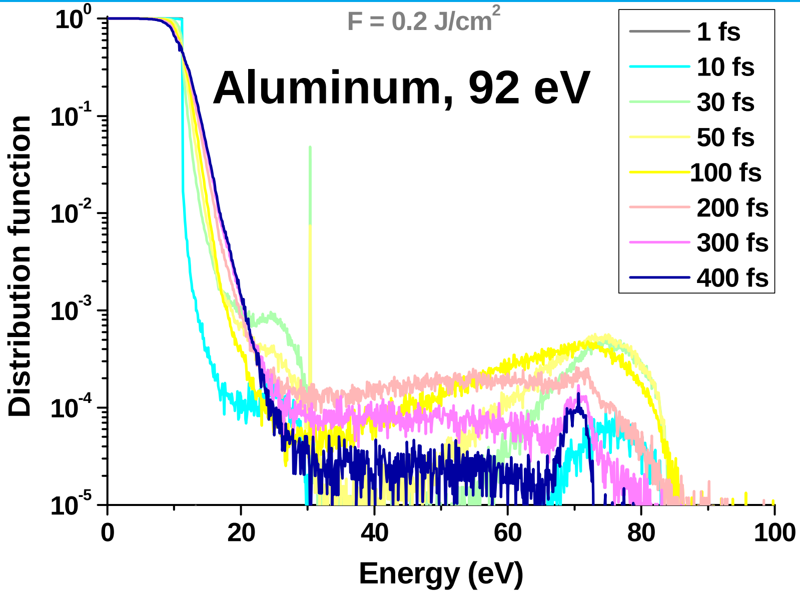


[N. Medvedev, B. Rethfeld, NJP **12** (2010)]

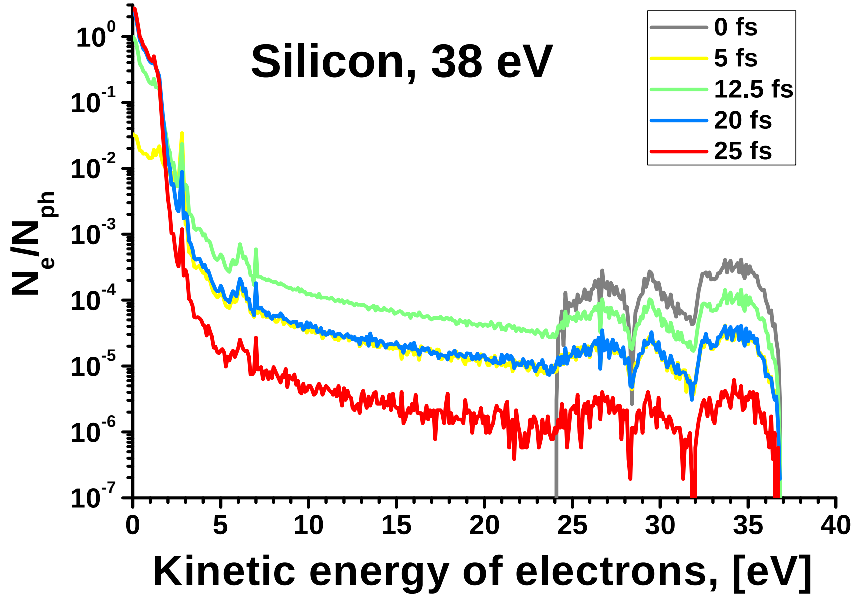
[R.R. Faustlin, B. Ziaja *et al.*, PRL **104** (2010)]



Electron distribution function



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



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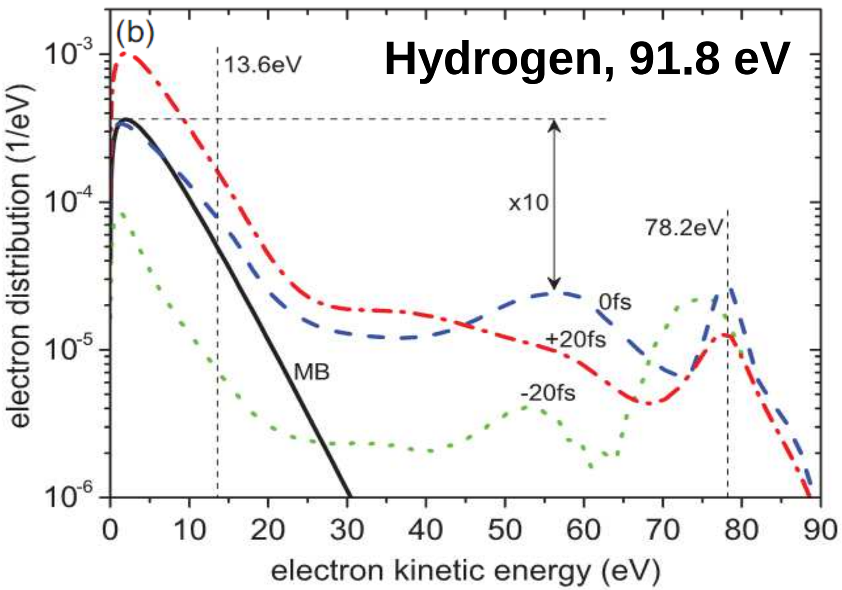
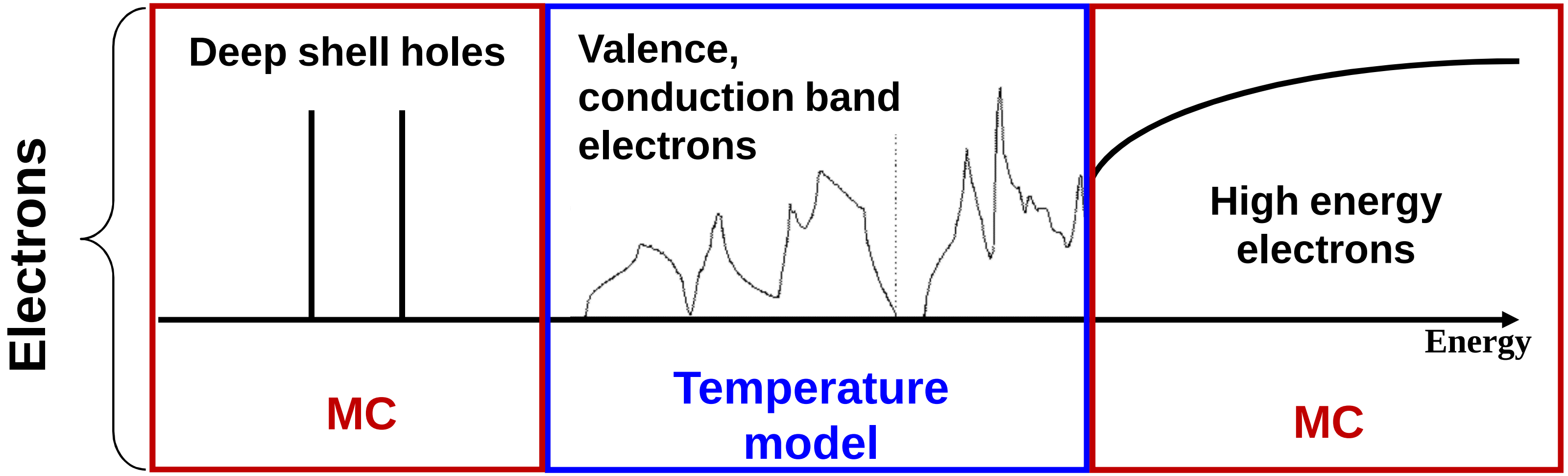


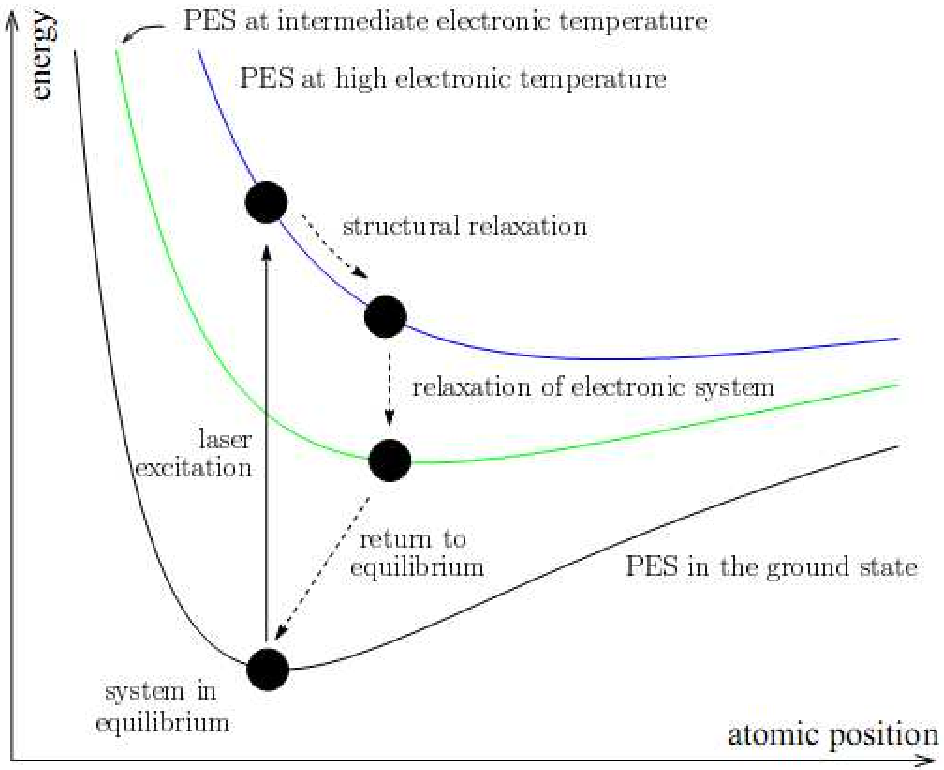
"Bump on hot tail" distribution:

- thermalized low energy part
- high energy non-thermalized tail

[D.Chapman, D. Gericke, PRL **107** (2011)]

Combined MC-TBMD





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B. Ziaja, N. Medvedev, HEDP **8**, 18 (2012)
N. Medvedev, H. Jeschke, B. Ziaja, NJP (*accepted*)

Processes considered

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) **Lattice heating, atomic dynamics**
6) **Changes of band structure**
7) Changes of scattering rates (not included yet)

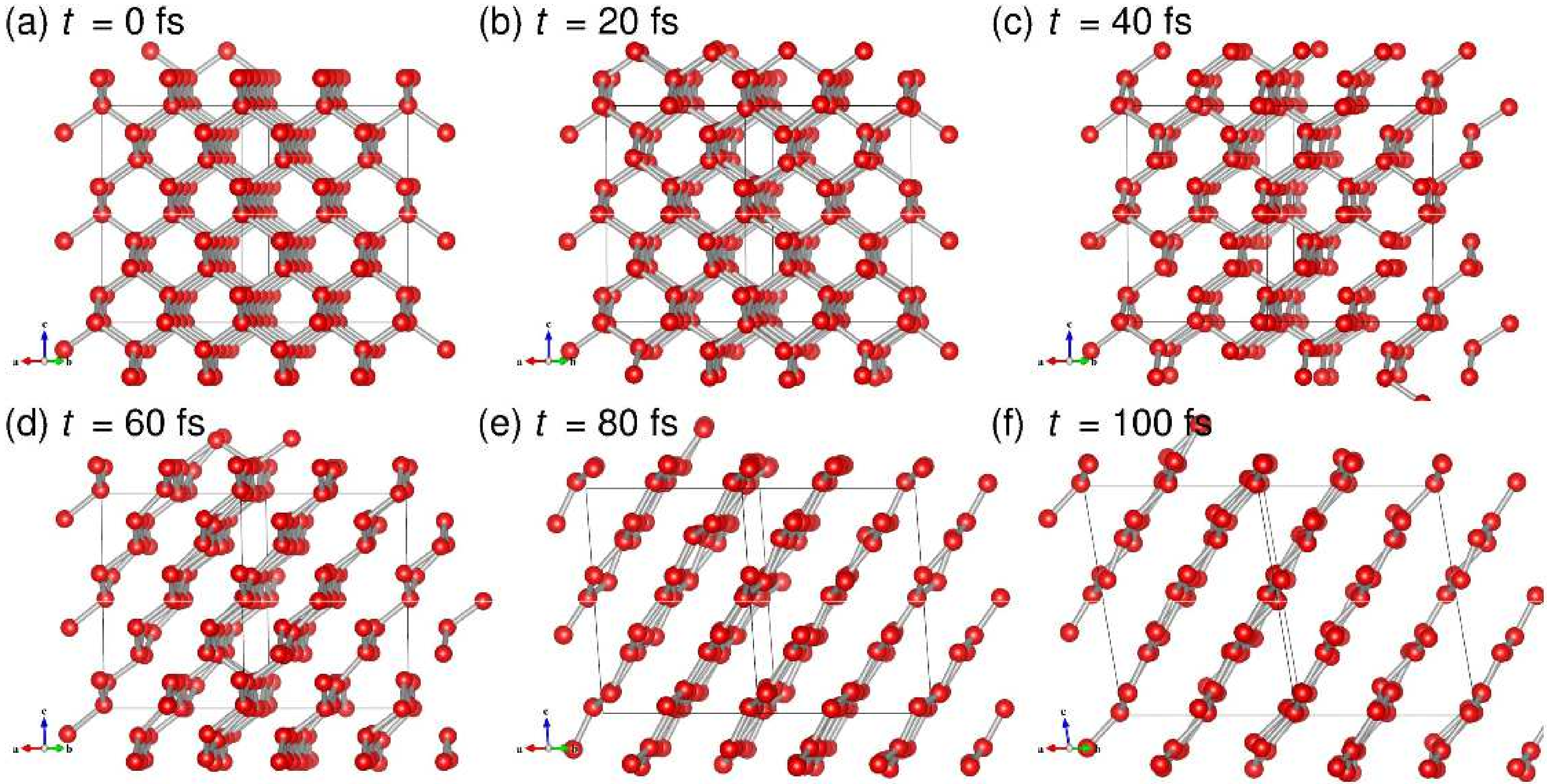
} **- TBMD**

N. Medvedev, H. Jeschke, B. Ziaja, NJP (*accepted*)

DESY

Results: Atomic motion

Photon energy 92 eV, FWHM = 10 fs

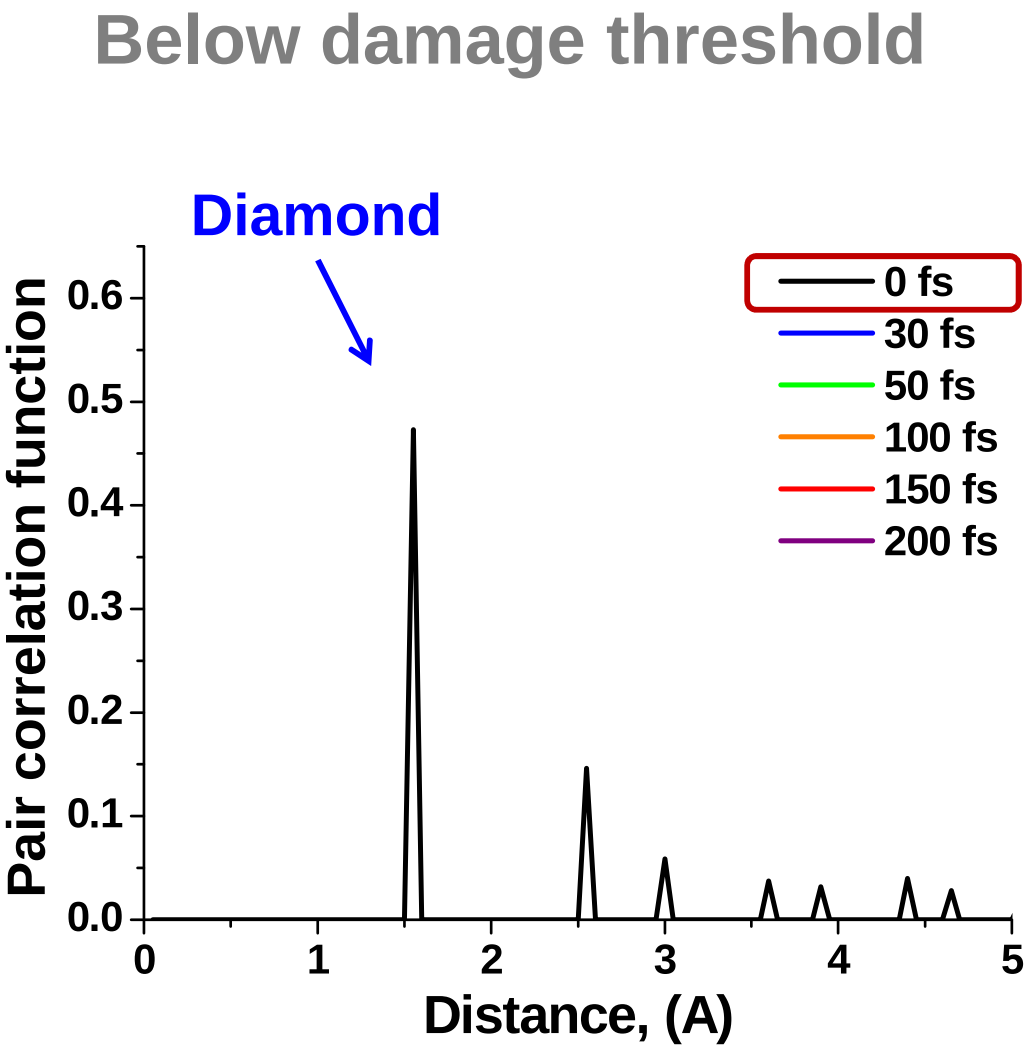


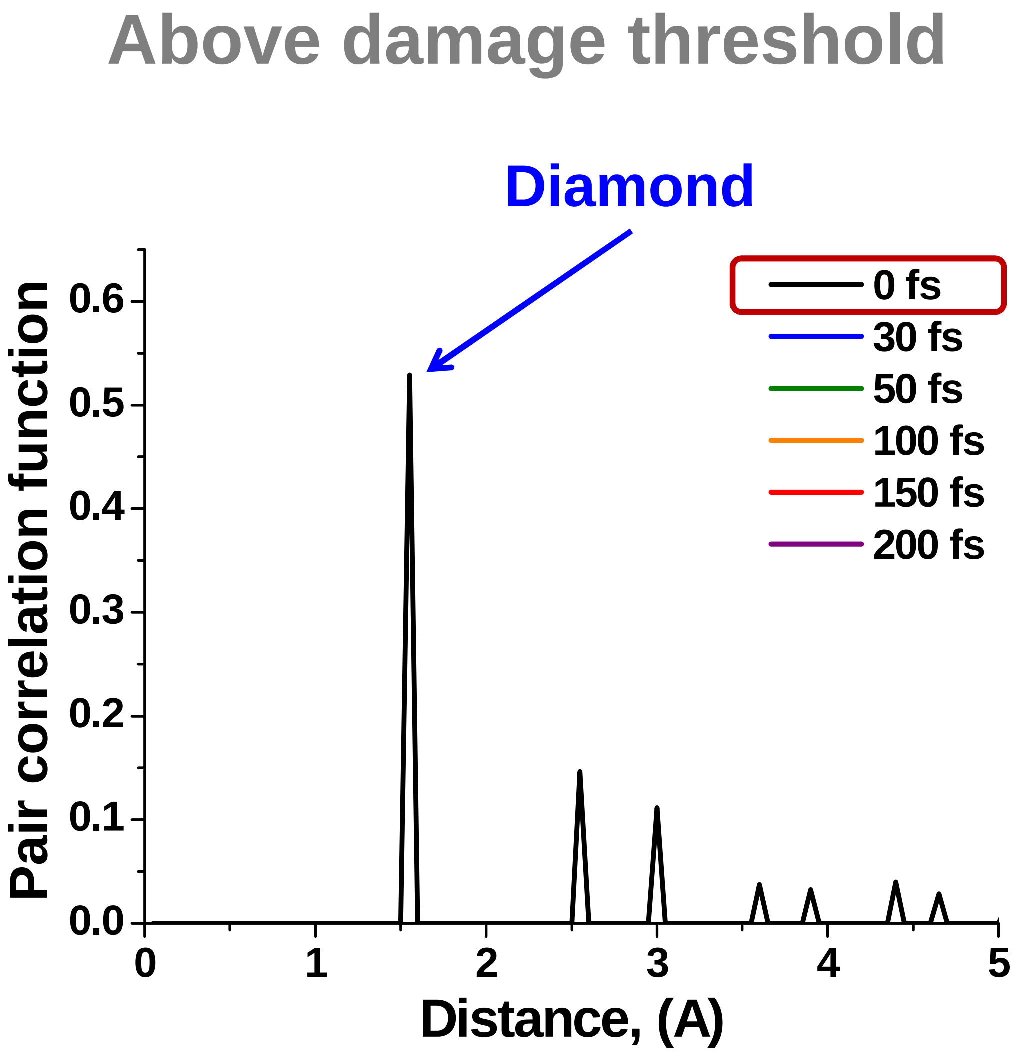
Ultrafast graphitization of diamond

N. Medvedev, H. Jeschke, B. Ziaja, NJP (*accepted*)

Results: Pair Correlation Function

Photon energy 92 eV, FWHM = 10 fs





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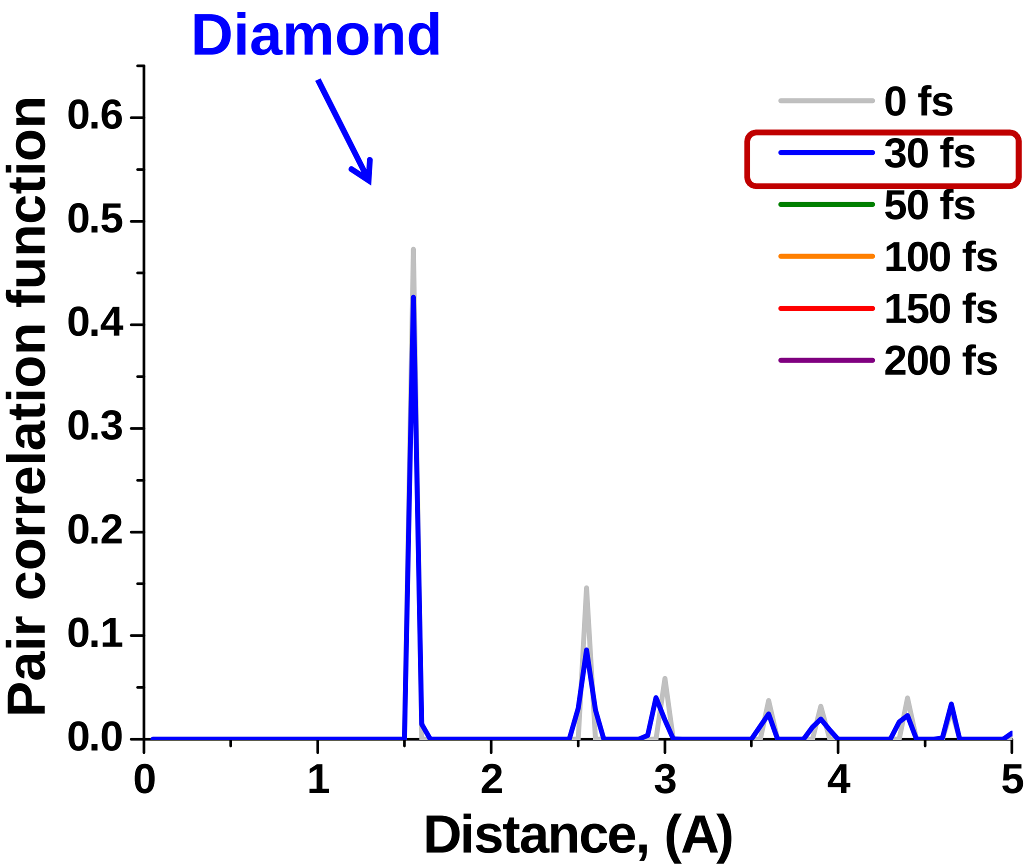
Below damage threshold



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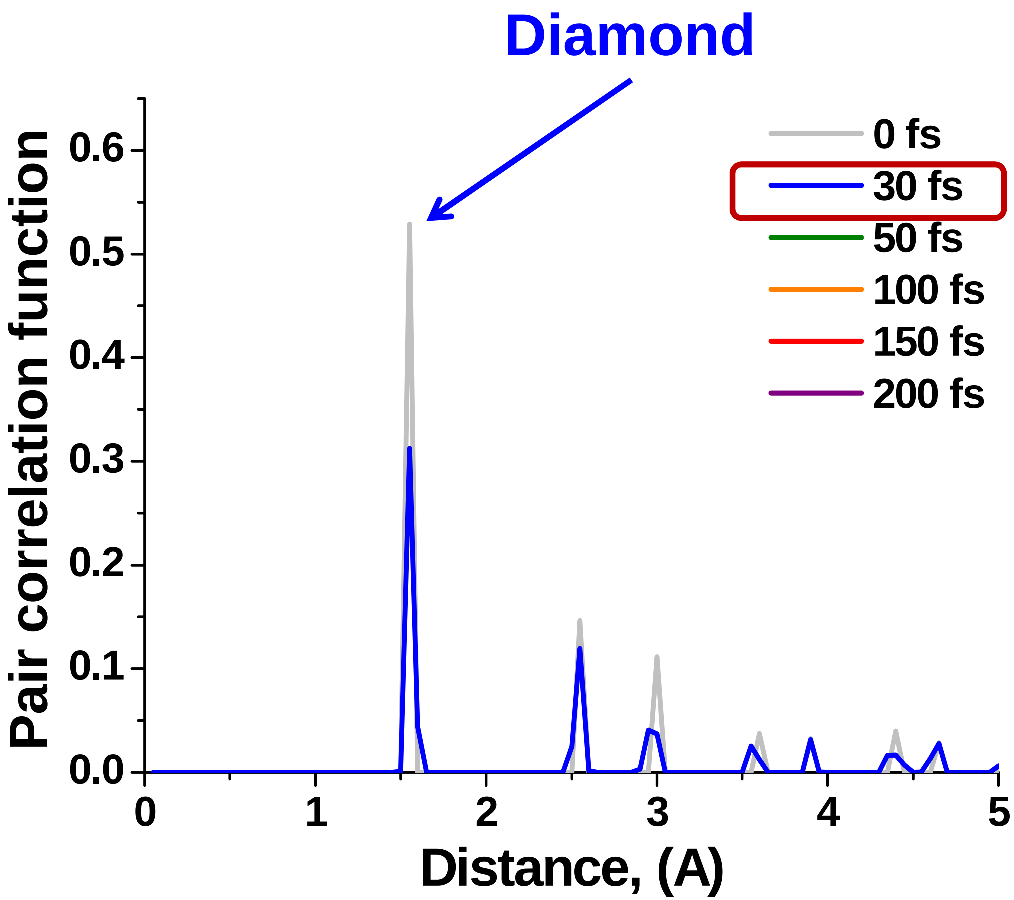

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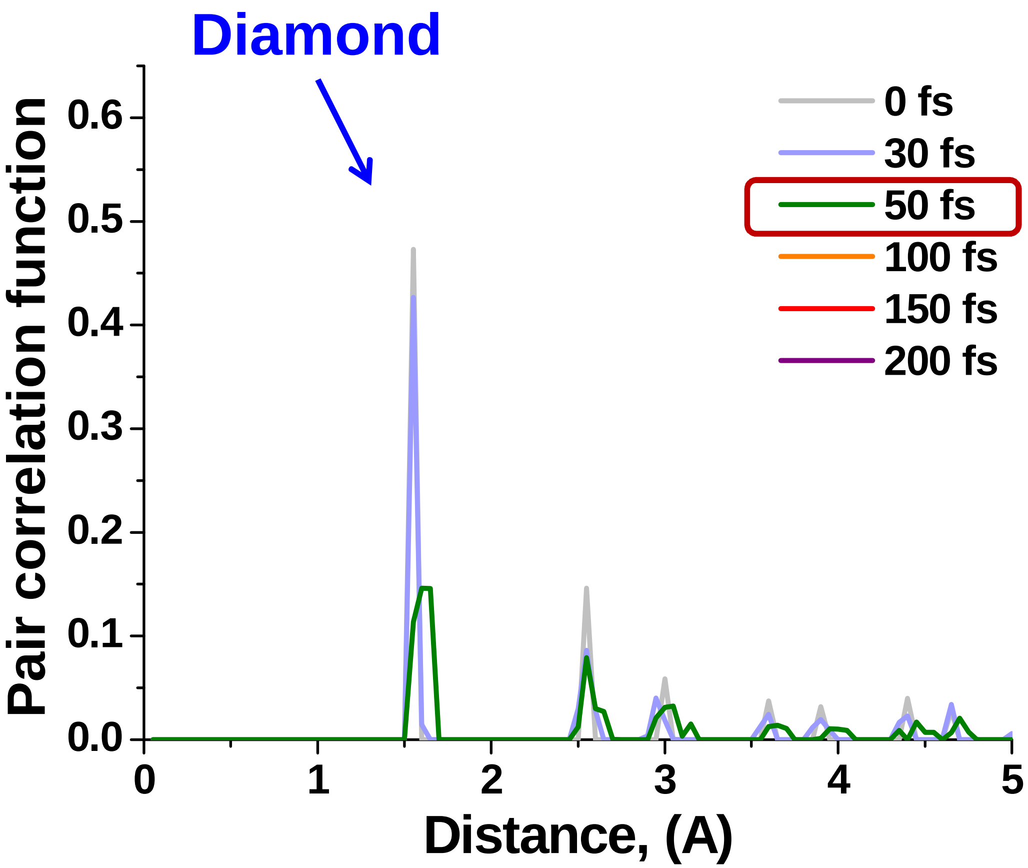

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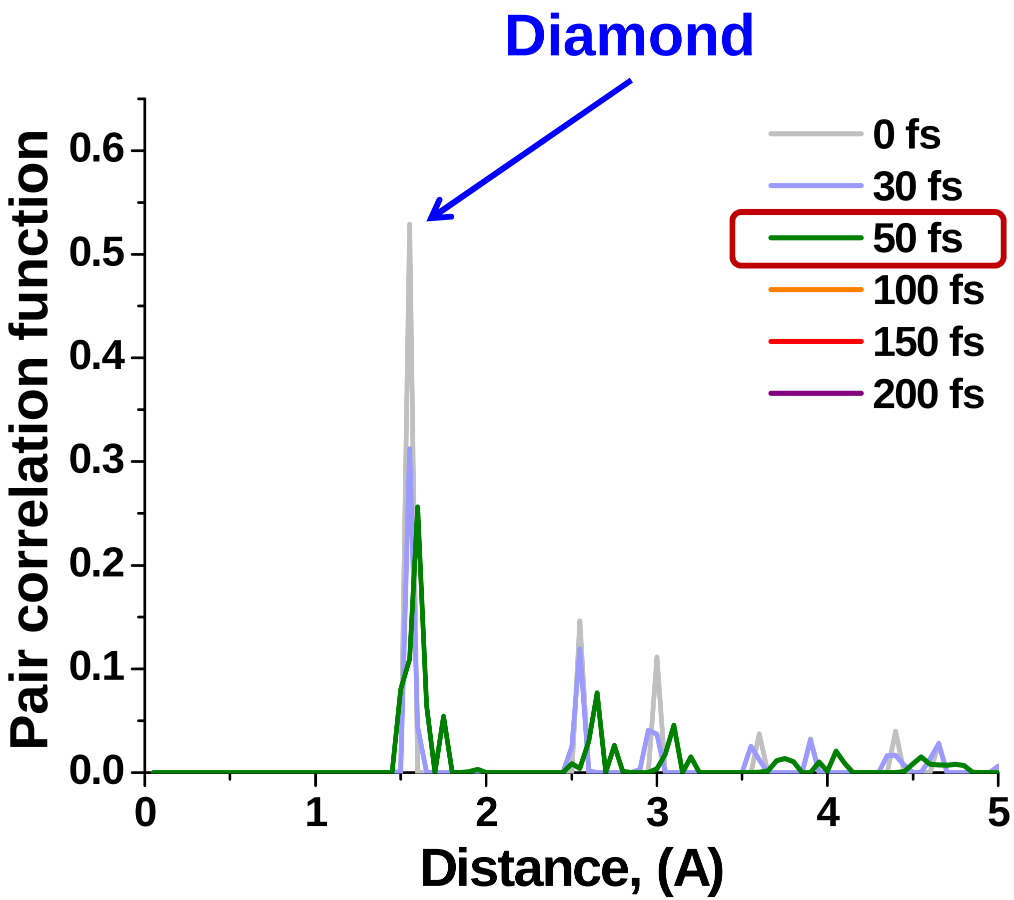

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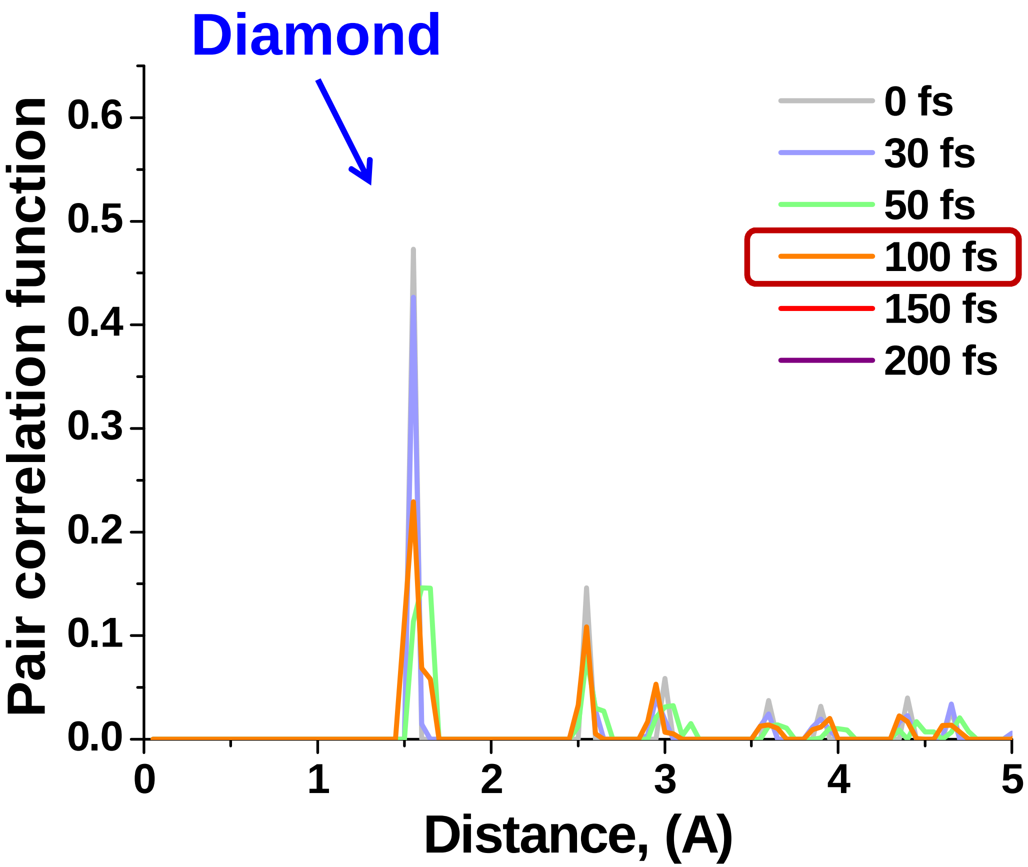

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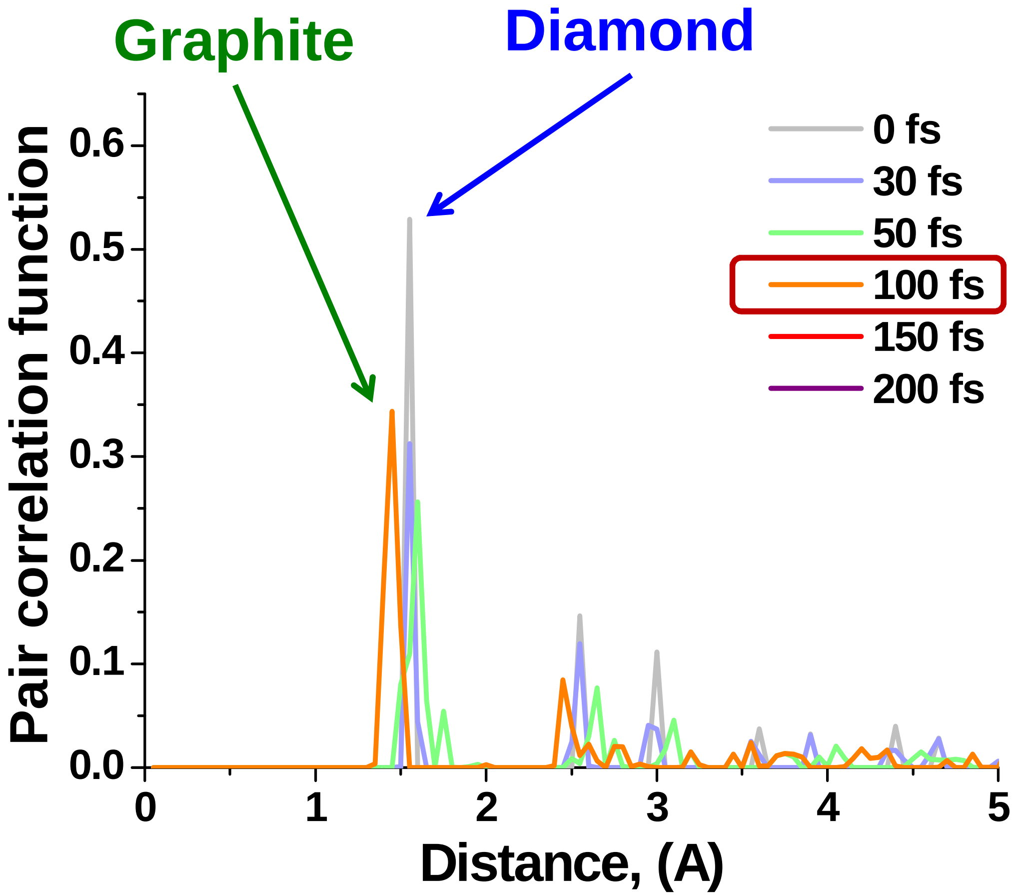

For higher fluence: ultrafast phase transition

DESY

Results: Pair Correlation Function

Photon energy 92 eV, FWHM = 10 fs

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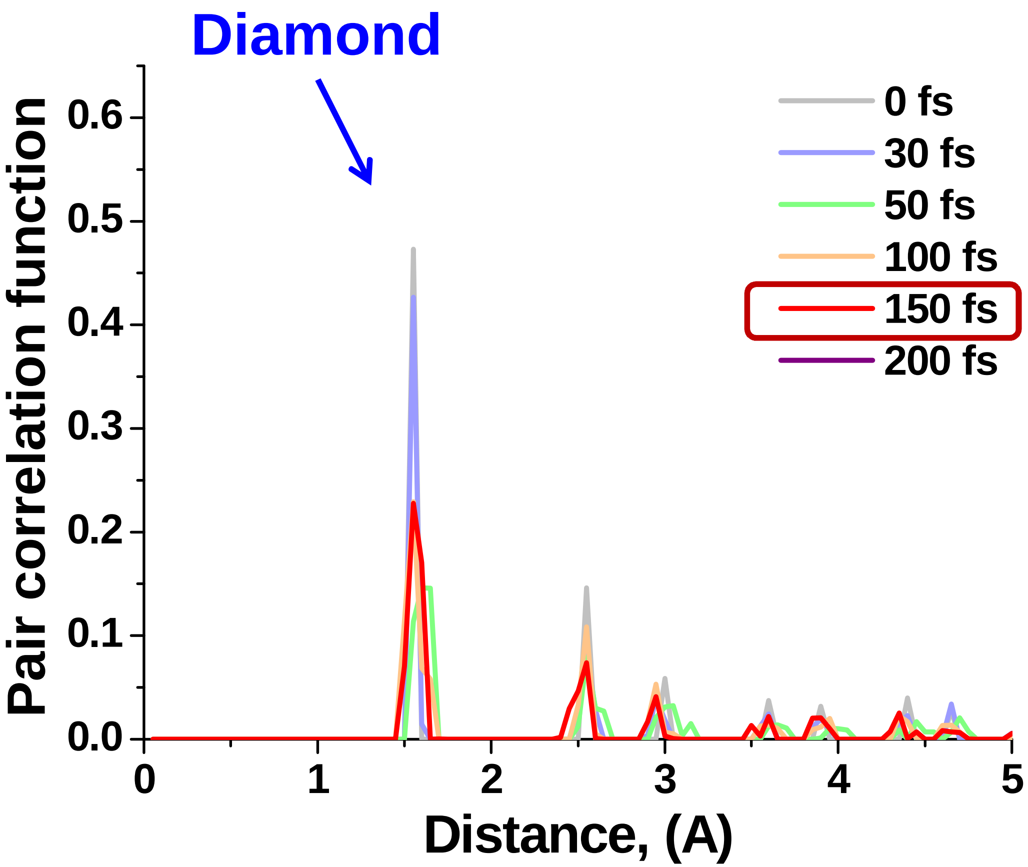

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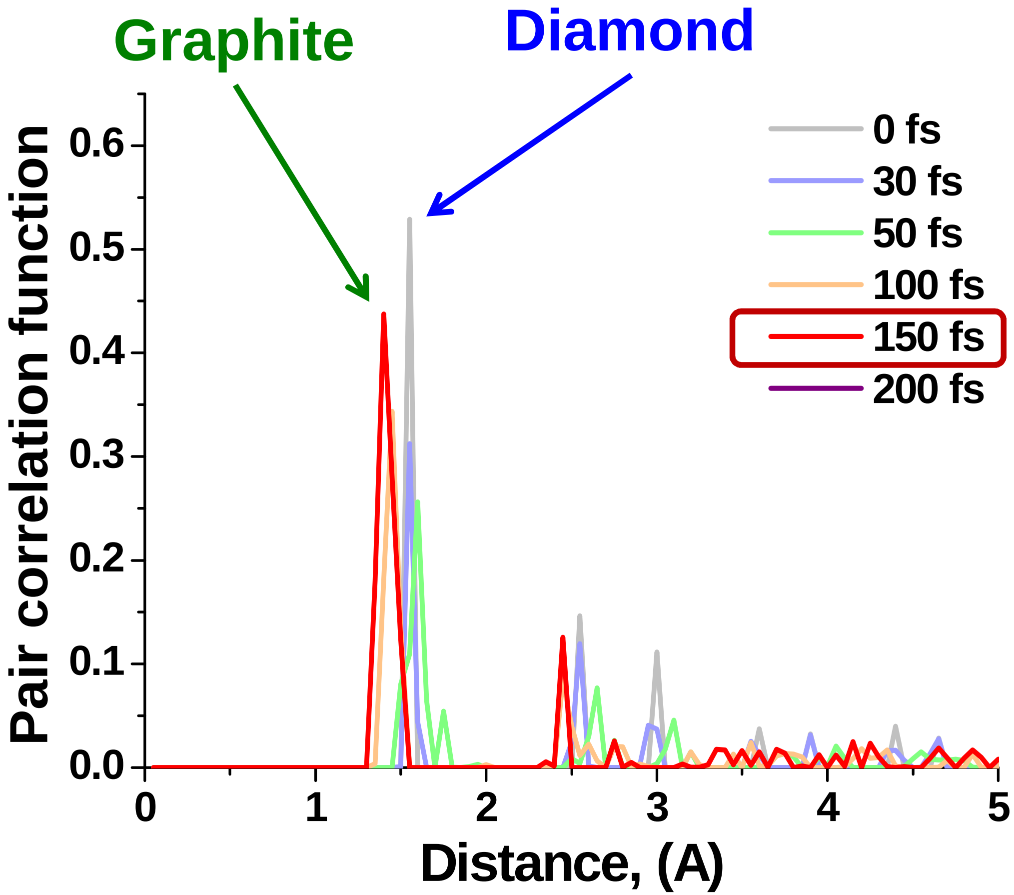

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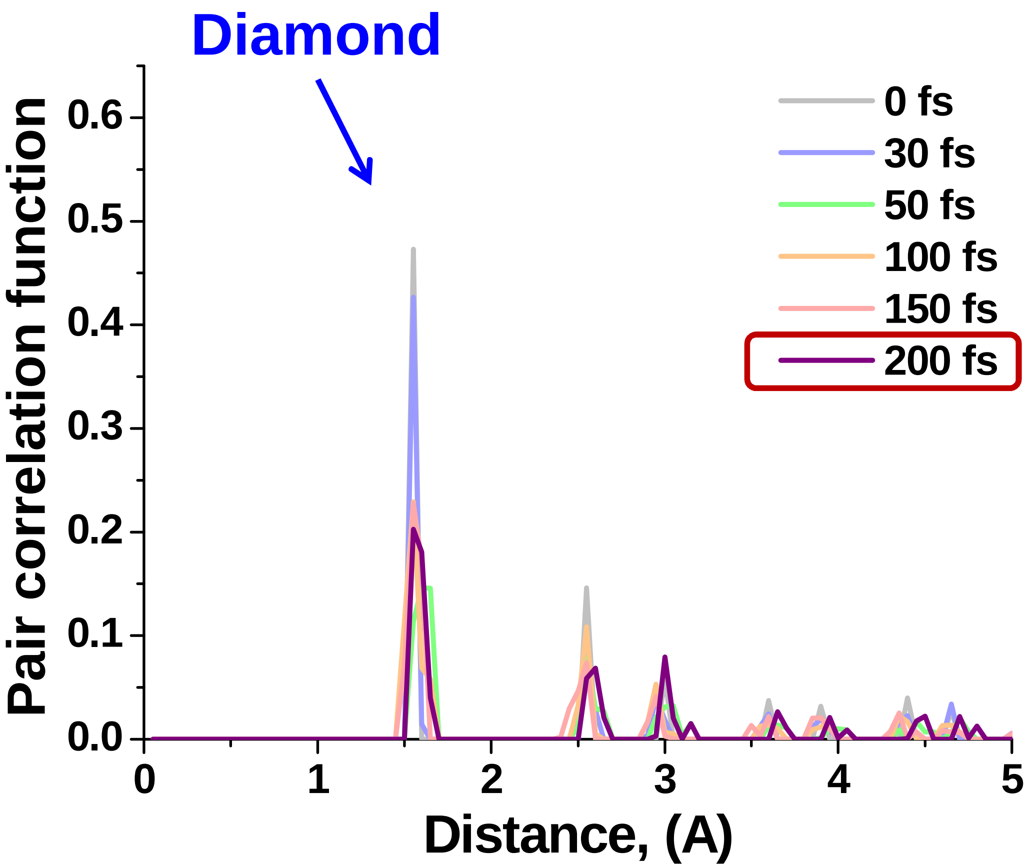
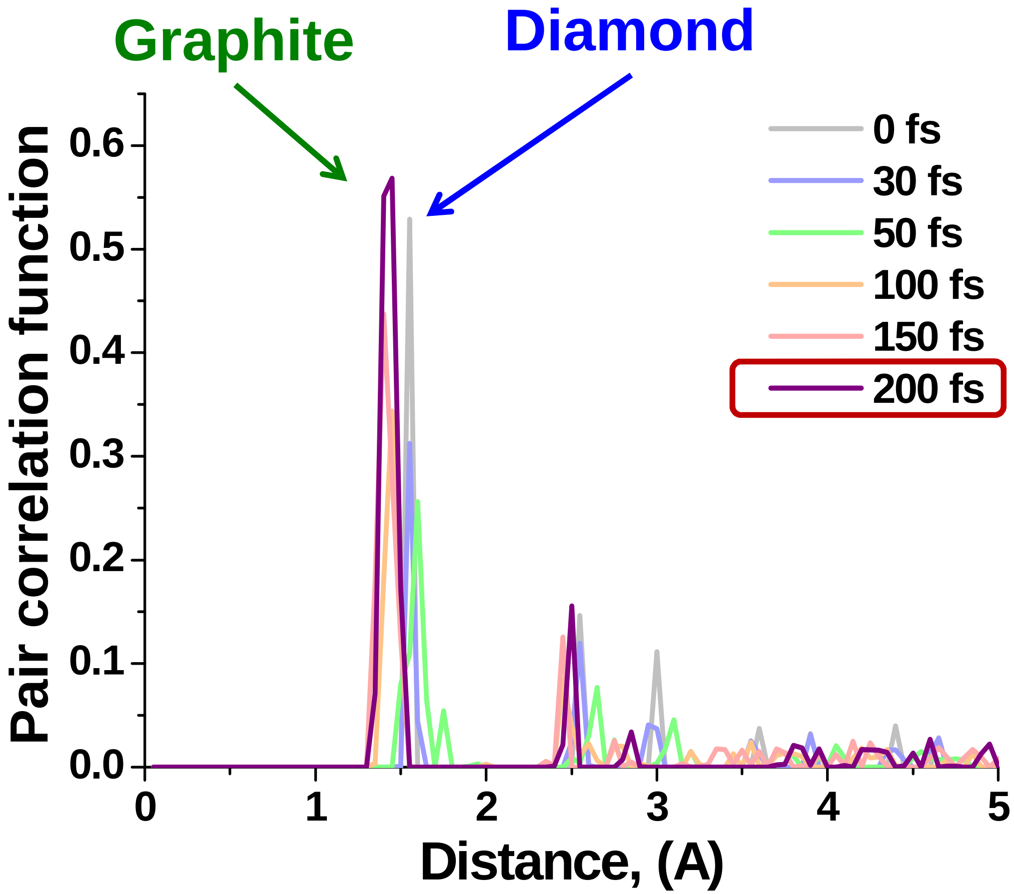


Above damage threshold



Damage threshold: 0.7 eV/atom (good agreement with experiments by J. Gaudin *et al.*)

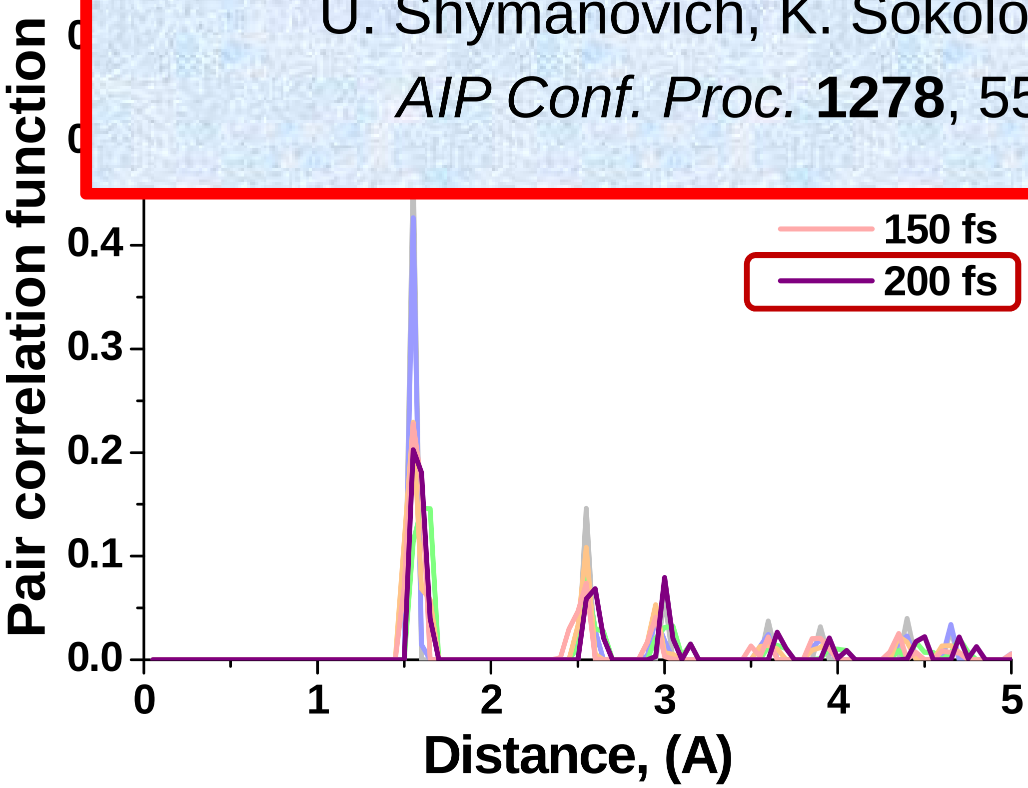
Results: Pair Correlation Function

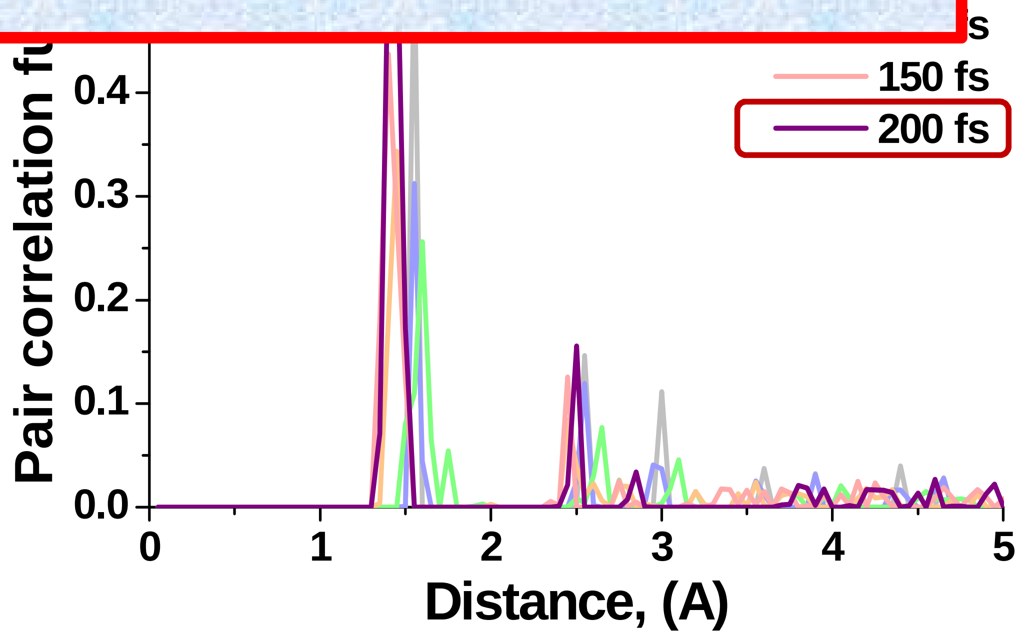
Experiments desirable:

Time resolved X-ray diffraction measuring phase transitions

S. P. Hau-Riege *et al, PRL* **108**, 217402 (2012)

U. Shymanovich, K. Sokolowski-Tinten *et al*,
AIP Conf. Proc. **1278**, 558 (2010)



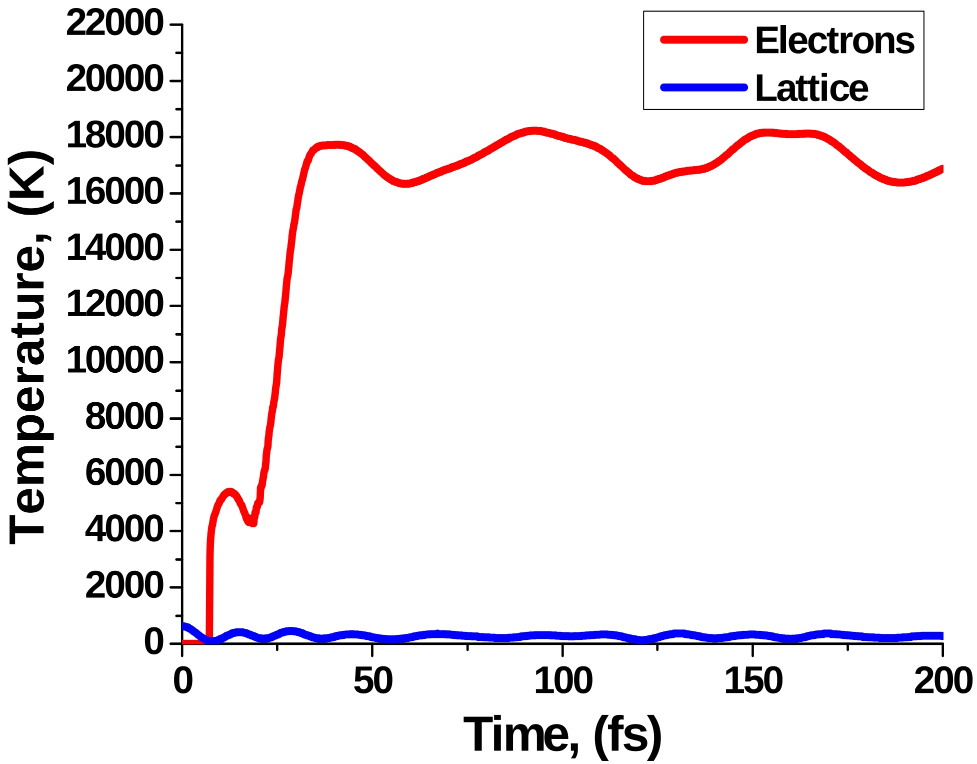


Damage threshold: 0.7 eV/atom (good agreement with experiments by J. Gaudin *et al.*)

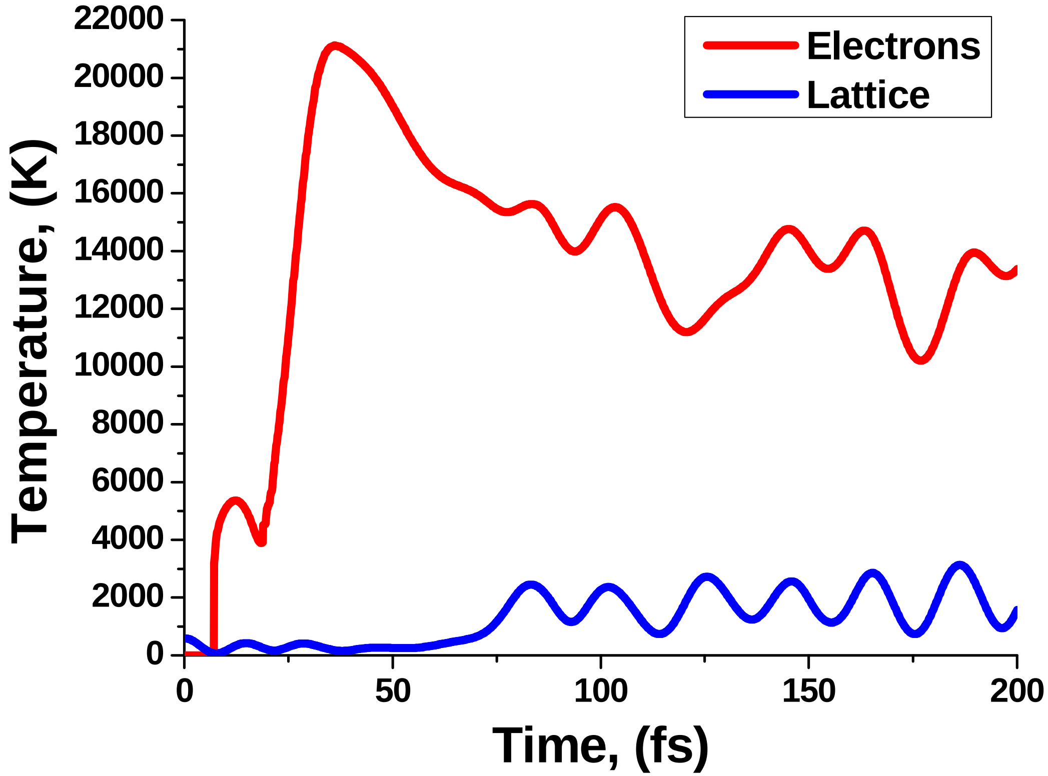
Results: Temperatures

Photon energy 92 eV, FWHM = 10 fs

Below damage threshold



Above damage threshold



Nonthermal phase transition

Damage threshold: 0.7 eV/atom (good agreement with experiments by J. Gaudin *et al.*)

Results: Temperatures

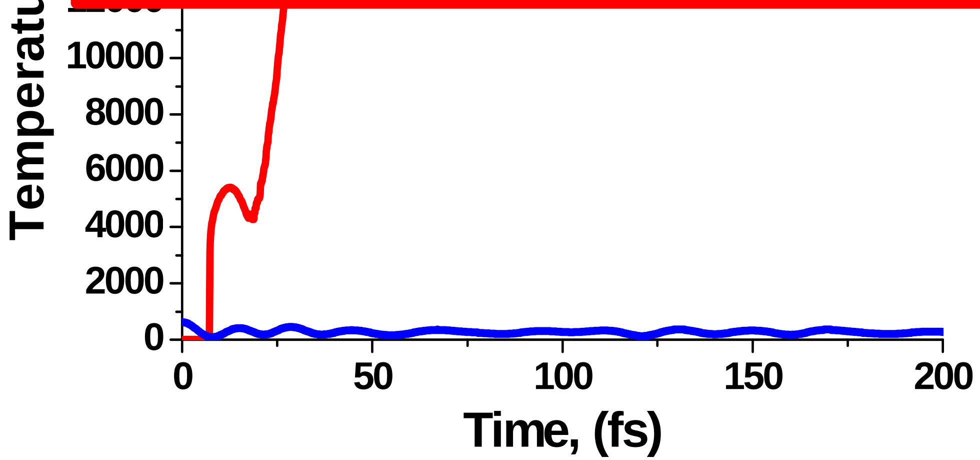
Photon energy 92 eV, FWHM = 10 fs

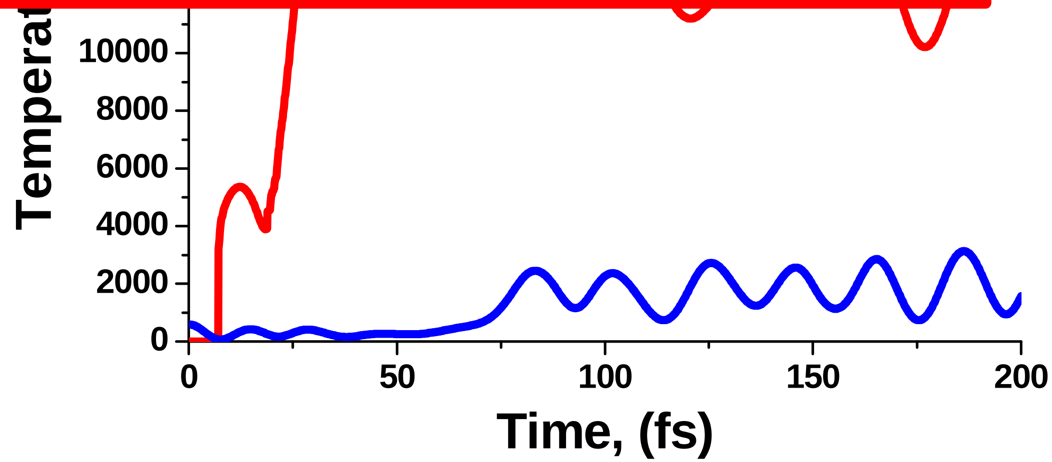
Experiments are highly desirable:

- **Time resolved atomic temperature (X-ray probe, Debye-Waller)**

  K. Sokolowski-Tinten *et al*, *Nature* **422**, 287 (2003)

- **Time resolved electron <u>and</u> atomic temperatures (combined visible light + X-ray probes?) – measuring energy flows**





Nonthermal phase transition

Damage threshold: 0.7 eV/atom (good agreement with experiments by J. Gaudin *et al.*)

Outlook

Future experiments should help to understand:

- Kinetic coefficients

optical coefficients, cross-sections, band-structure of excited matter

- Transient electron kinetics

electron density, temperature, distribution

- Atomic kinetics

structural changes, timescales

- Simultaneous electrons-atoms kinetics

electron and atom temperatures, electron-phonon coupling, energy exchange rate

Thank you for your attention!

Thanking collaborators:

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