Modeling medium photo-excitation of condensed matter and its structural changes

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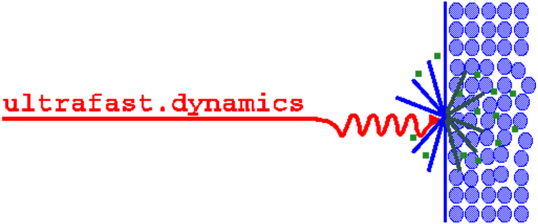
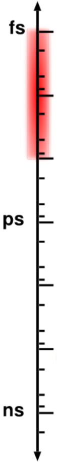


Time scales of laser-induced processes



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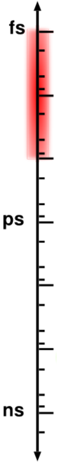
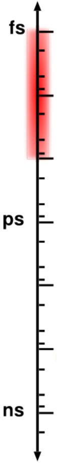


Electron excitation ...

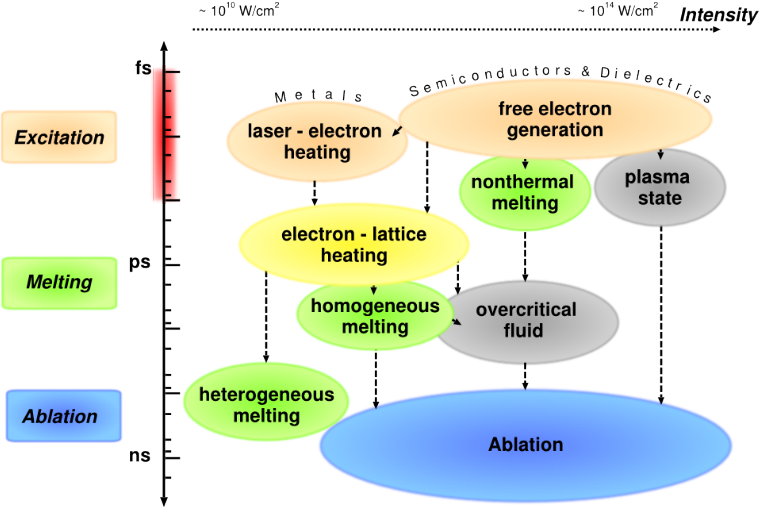
Time scales of laser-induced processes



Electron excitation ...

... induces phase transition

Time scales of laser-induced processes



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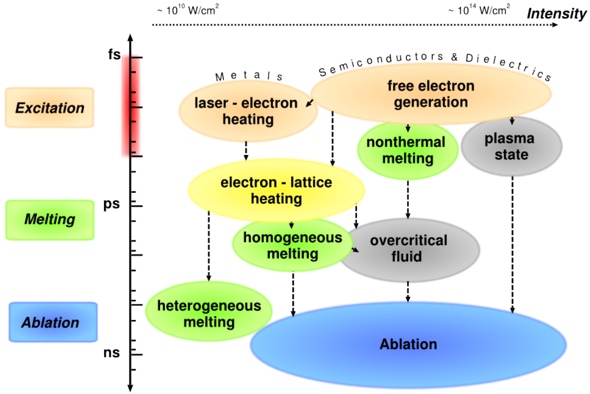

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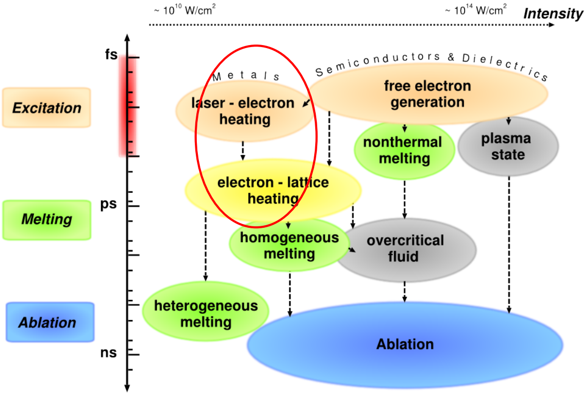
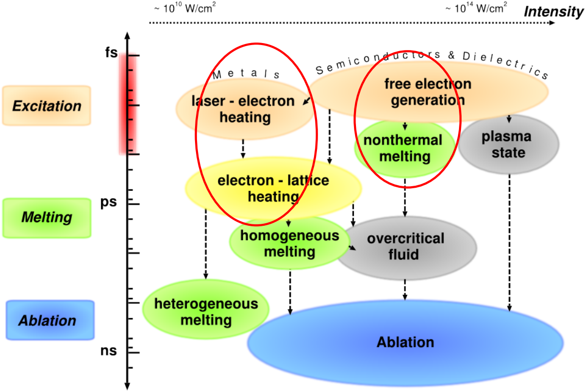


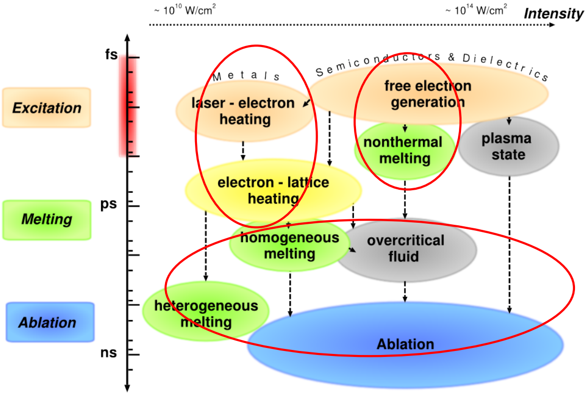
- Energy distribution

Time scales of laser-induced processes



- Energy distribution
- Electron density

Time scales of laser-induced processes



- Energy distribution
- Electron density
- Structural dynamics

Main questions

- How does the energy distribution look like?

→ Photo-emission experiments

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- How many electrons are excited?

→ Density dependence of reflectivity $R(t)$

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- How does the energy distribution look like?

→ Photo-emission experiments

- How many electrons are excited?

→ Density dependence of reflectivity $R(t)$

- Which structural changes are induced?
- How fast they occur?

→ Time-resolved X-ray probing

Idea and structure of the talk

Calculate energy distribution

(methods: Boltzmann collision terms, Monte Carlo simulation)

↪ evolution of electron density

↪ energy transfer to lattice/atoms

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1. Visible light

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1. Visible light
2. XUV irradiation

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1. Visible light
2. XUV irradiation $\Longrightarrow$
   - Water
   - Silicon
   - Aluminum

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Calculate energy distribution

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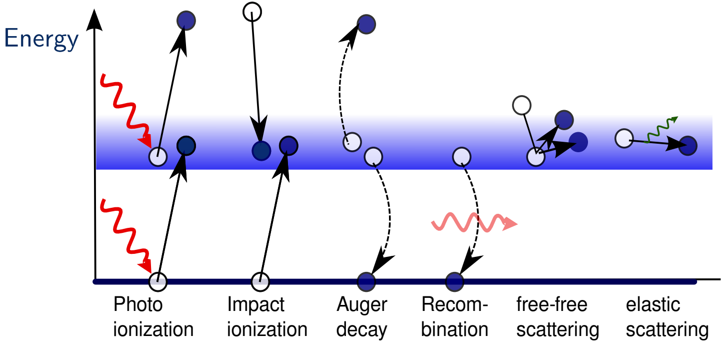
1. Visible light
2. XUV irradiation ⟹
   - Water
   - Silicon
   - Aluminum

Hybrid simulation including lattice dynamics

3. Example for visible light

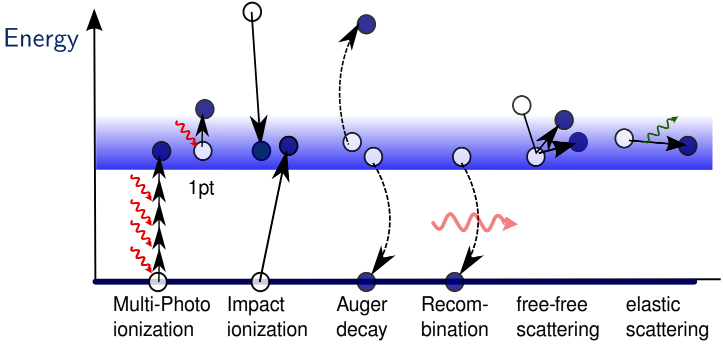
XUV or visible light

Which processes change energy and density of free electrons?

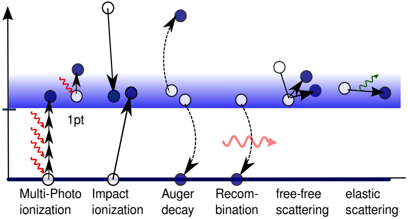


XUV or visible light

Which processes change energy and density of free electrons?
Difference for visible light



1. Visible light



[1] Kaiser, Rethfeld et al., Physical Review B **61**, 11437 (2000)

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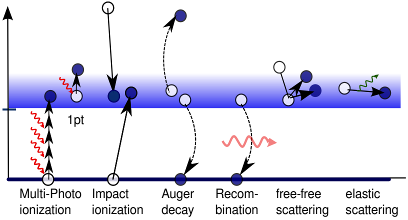

$$\frac{\partial f(\mathbf{k})}{\partial t} = \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{el-el}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{el-phon}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{Laser1pt}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{MPI}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{ImpIonis}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{AugerRec}}$$

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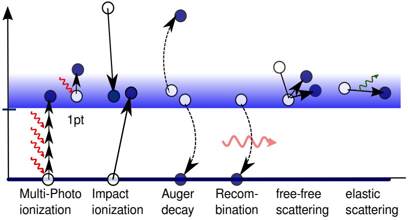

New compared to [1]:

- Auger recombination
- Above-threshold ionization
- Valence band dynamics
- Density-dependent optical parameters

$$\frac{\partial f(\mathbf{k})}{\partial t} = \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{el-el}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{el-phon}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{Laser1pt}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{MPI}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{ImpIonis}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{AugerRec}}$$

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1. Visible light

Change of electrons distribution function

$$\frac{\partial f(\mathbf{k})}{\partial t} = \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{el-el}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{el-phon}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{Laser1pt}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{MPI}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{ImpIonis}} + \left(\frac{\partial f(\mathbf{k})}{\partial t}\right)_{\mathrm{AugerRec}}$$

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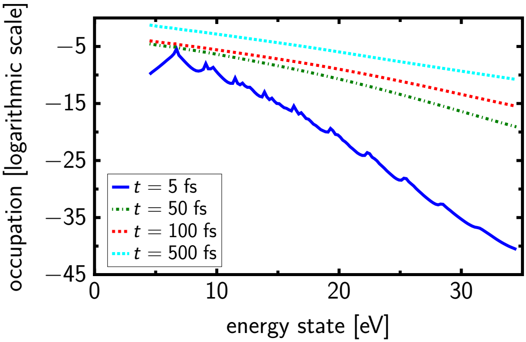
Each process described by a complete Boltzmann collision integral

$$\frac{\partial f(\mathbf{k})}{\partial t} = \sum_{\substack{\text{all } \mathbf{k'} \\ \text{of collision partners}}} M^2(\mathbf{k},\mathbf{k}') \times \mathcal{F}[f(\mathbf{k}), f(\mathbf{k}'), f(\mathbf{k}\pm\mathbf{k}')] \times \delta(\varepsilon(\mathbf{k}) - \varepsilon(\mathbf{k}\pm\mathbf{k}'))$$

$$= \text{probability of collision} \times \text{Pauli's principle} \times \text{energy conservation}$$

1. Visible light

Transient electron distribution function

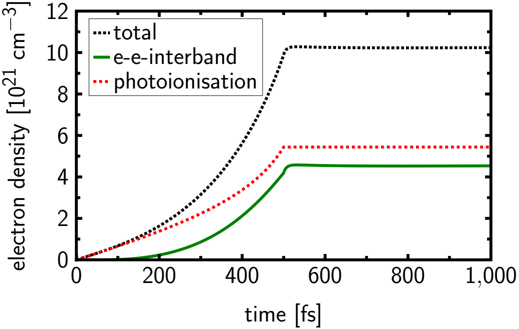


- Parameters for SiO_2
- Constant laser intensity
- Photon energy $\hbar\omega_L = 2.5\ \mathrm{eV}$

- Excitation to low energies by multiphoton ionization
- Intraband absorption repeats peaks
- Increase of distribution due to further ionization

1. Visible light

Resulting electron density



Pulse duration 500 fs

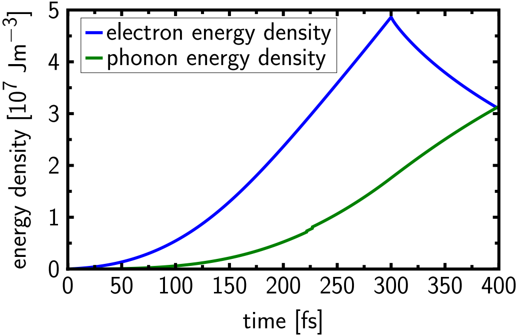
$$n_e(t) =$$

$$\Omega \int dE\, D(E) \cdot f(E,t)$$

- Linear increase due to photoionization, deviation due to changing optical parameters
- Interband processes add net electrons

1. Visible light

Energy transfer to phonons



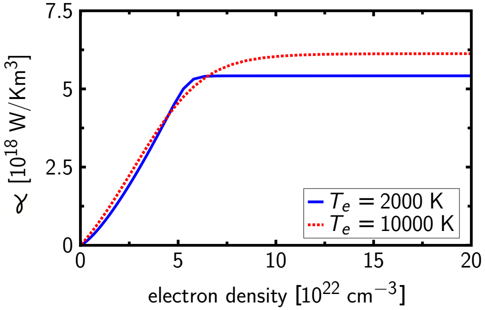
$$u_e(t) = \Omega \int dE\, D(E) \cdot f(E,t)\; E$$

$$u_{\mathrm{ph}}(t) = \Omega \int dE\, D(E) \cdot g(E,t)\; E$$

Electron energy	increases due to laser excitation
	decreases due to phonon emission
Phonon energy	increases during the pulse
	larger than electron energy when thermalized

1. Visible light

Energy transfer to phonons



Extract parameter for
two-temperature description

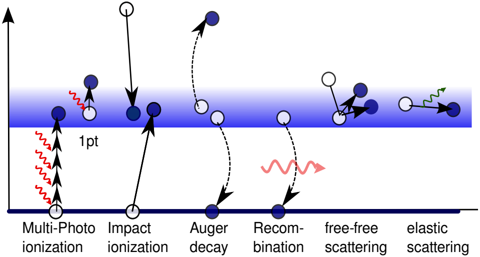
$$\frac{\partial u_e}{\partial t} = -\alpha\,(T_e - T_{\mathrm{ph}})$$

$$\frac{\partial u_{\mathrm{ph}}}{\partial t} = +\alpha\,(T_e - T_{\mathrm{ph}})$$

- linear increase with density
- asymptotic behaviour for degenerate electrons
- slightly depending on electron temperature
- **Idea:** include in two–temperature heat conduction model for dielectrics

2. XUV irradiation

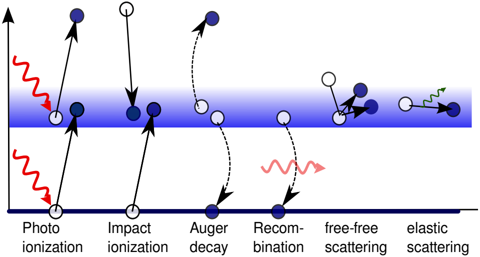
Which processes change energy of electrons?
Visible light



2. XUV irradiation

Which processes change energy of electrons?

XUV irradiation



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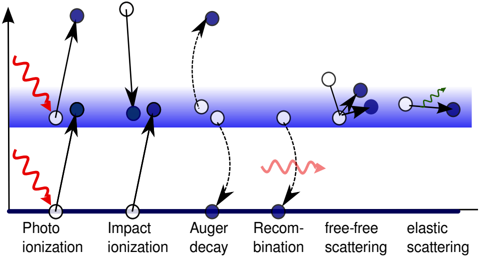
Which processes change energy of electrons?
XUV irradiation



Monte Carlo simulation:

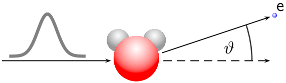
- trace each electron event by event
- decide for each timestep which process will happen
- random number determines its particular realization

2. XUV irradiation

Excitation of liquid water

Currently included processes:

- photoionization



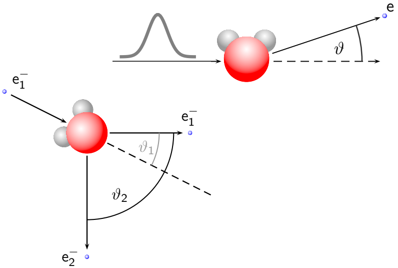
2. XUV irradiation

Excitation of liquid water

Currently included processes:

- photoionization

- secondary ionization

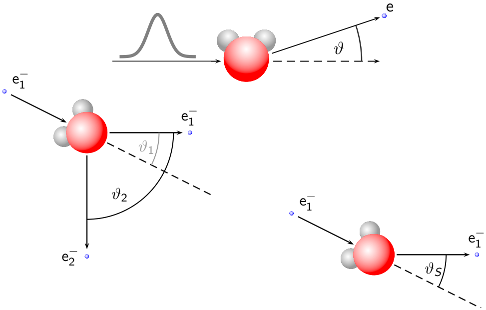


2. XUV irradiation

Excitation of liquid water

Currently included processes:

- photoionization
- secondary ionization
- elastic scattering

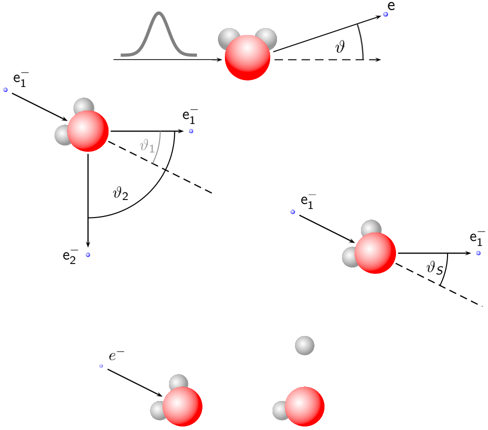


2. XUV irradiation

Excitation of liquid water

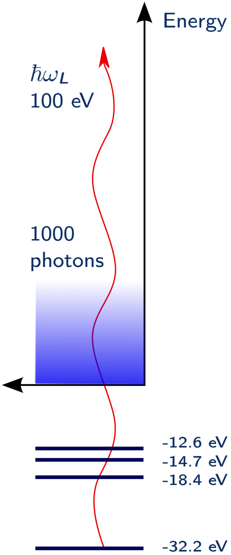
Currently included processes:

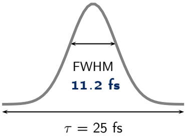
- photoionization
- secondary ionization
- elastic scattering
- recombination



2. XUV excitation of water

Transient energy distribution





2. XUV excitation of water

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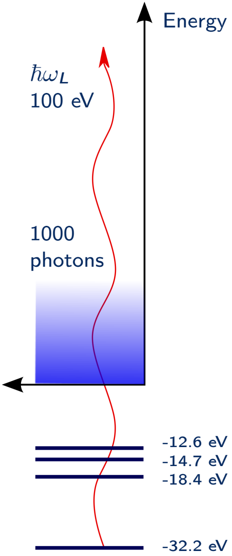
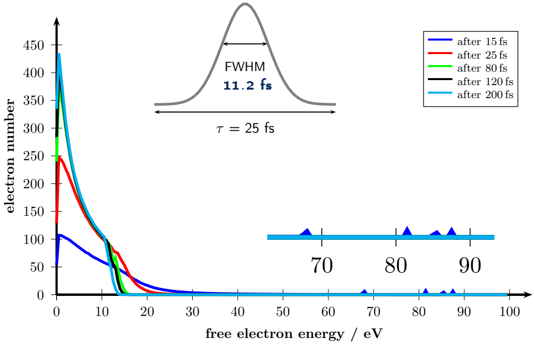

Energy

$\hbar\omega_L$
100 eV

1000
photons

-12.6 eV
-14.7 eV
-18.4 eV

-32.2 eV



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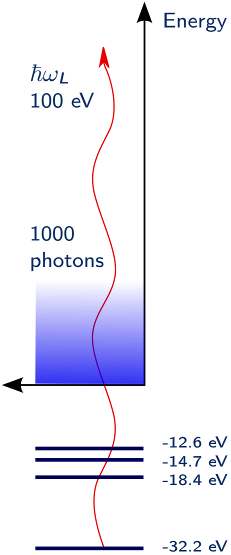
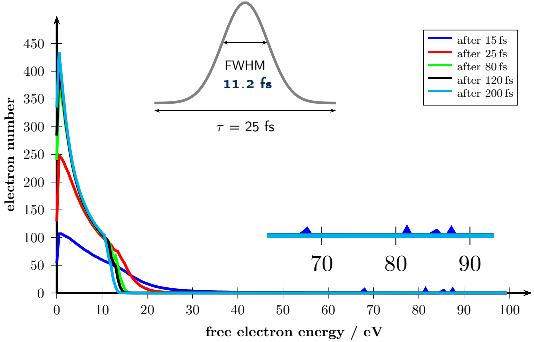
Transient energy distribution

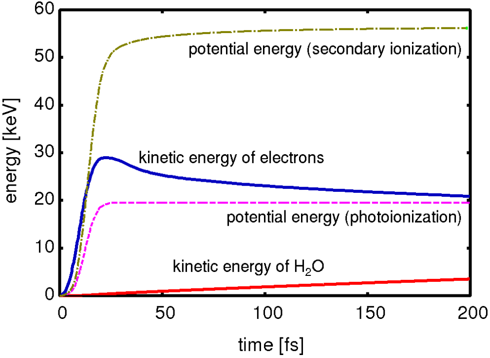




- Initial peaks decay quickly
- No secondary ionization for energies below 12.6 eV
- Distribution shift due to elastic collisions

2. XUV excitation of water

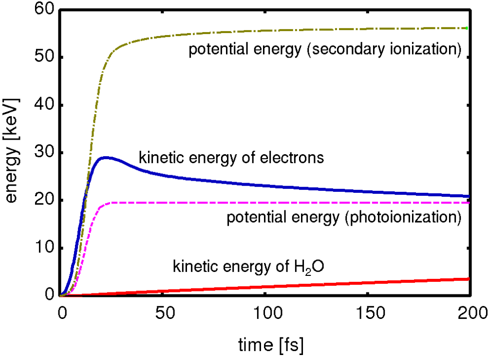
Excitation and energy balance



- Potential energy of electron-hole pairs
- Kinetic energy of electrons or molecules

2. XUV excitation of water

Excitation and energy balance



- Potential energy of electron-hole pairs
- Kinetic energy of electrons or molecules

- Secondary ionization continues for ≈ 200 fs
- Energy transfer from electrons to H_2O molecules due to elastic scattering

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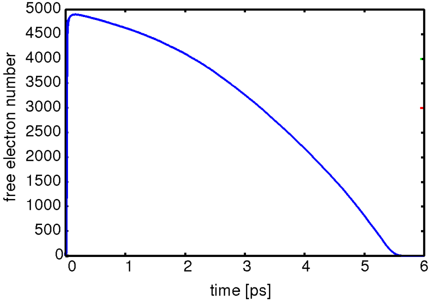
2. XUV excitation of water

Recombination

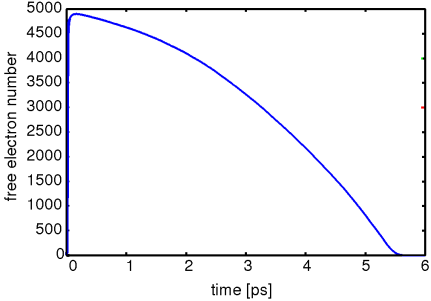
Electron decay



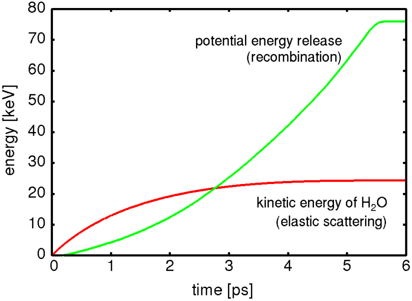
2. XUV excitation of water

Recombination

Electron decay



Energy of H_2O



2. XUV excitation of water

Recombination

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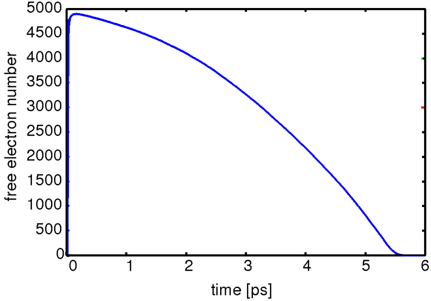

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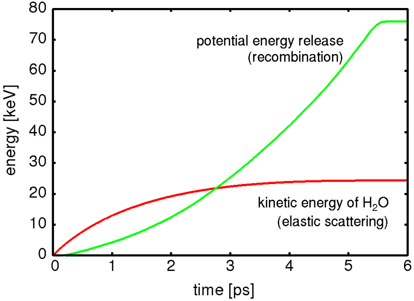

- Recombination process completed in the picosecond range
- Energy release due to recombination exceeds elastic scattering
- **Idea** to simulate subsequent Molecular Dynamics

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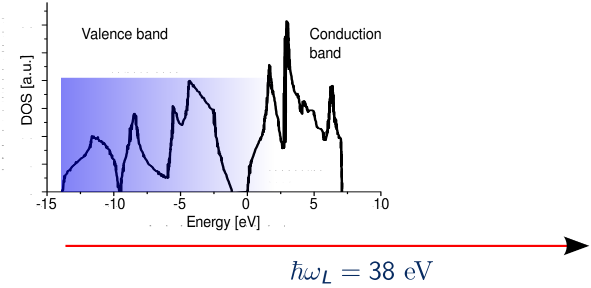
2. XUV irradiation

Excitation of silicon

Consider valence band dynamics



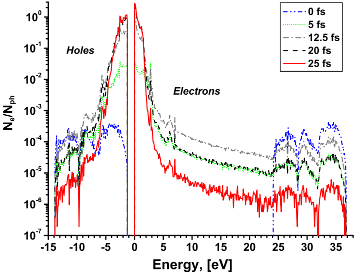
$\hbar\omega_L = 38$ eV

How many electrons finally excited?

Rough estimation through band gap: $N_{\mathrm{el}} \approx \dfrac{\hbar\omega_L}{E_{\mathrm{gap}}}$?

2. XUV excitation of silicon

Electron and hole dynamics

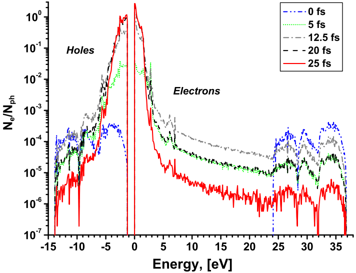


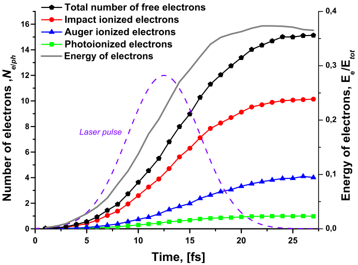
- Transient energy distribution of electrons and holes
- Electrons end mainly at low energies, holes at high energies

Medvedev and Rethfeld, New Journal of Physics **12** 073037 (2010)

2. XUV excitation of silicon

Electron and hole dynamics





- Transient energy distribution of electrons and holes
- Electrons end mainly at low energies, holes at high energies
- Electron decay due to impact ionization, hole raising by Auger-like process
- Ionization of secondary free electrons completed shortly after pulse

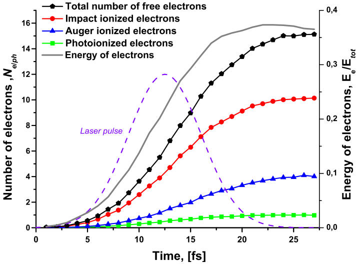
Medvedev and Rethfeld, New Journal of Physics **12** 073037 (2010)

2. XUV excitation of silicon

Effective energy gap

Each photon excites
$N_{\mathrm{el/pt}} \approx 15$ electrons

Compare with $\dfrac{\hbar\omega_L}{E_{\mathrm{gap}}} \approx 32.6$



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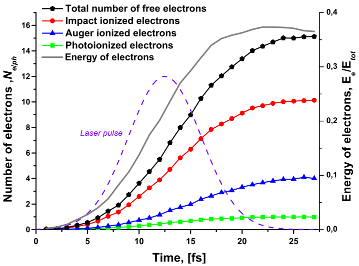
Compare with $\dfrac{\hbar\omega_L}{E_{\mathrm{gap}}} \approx 32.6$

$\Rightarrow$ Effective energy gap of
$E_{\mathrm{EEG}} = \hbar\omega_L / N_{\mathrm{el/pt}} = 2.6\ \mathrm{eV}$



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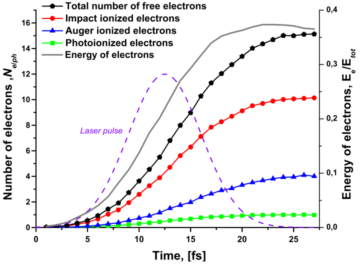


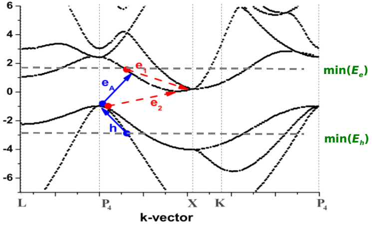
Excitation from any state of valence band to any state of conduction band

Effective energy gap is the statistically needed energy for pair creation

2. XUV excitation of silicon

Effective energy gap

Effective energy gap is the statistically needed energy for pair creation

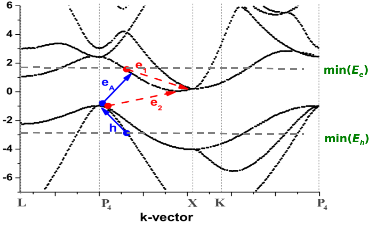


- Impact ionization
  needs minimum electron energy
- Auger ionization
  needs minimum hole energy
- Mean energy after ionization
  $\langle E_e \rangle = \frac{1}{2}\min(E_e)$, $\langle E_h \rangle = \frac{1}{2}\min(E_h)$

2. XUV excitation of silicon

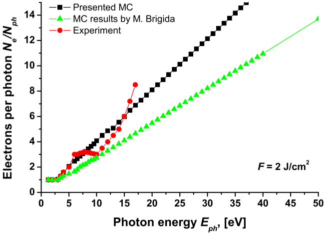
Effective energy gap

Effective energy gap is the statistically needed energy for pair creation



- Impact ionization
  needs minimum electron energy
- Auger ionization
  needs minimum hole energy
- Mean energy after ionization
  $\langle E_e \rangle = \frac{1}{2}\min(E_e)$, $\langle E_h \rangle = \frac{1}{2}\min(E_h)$

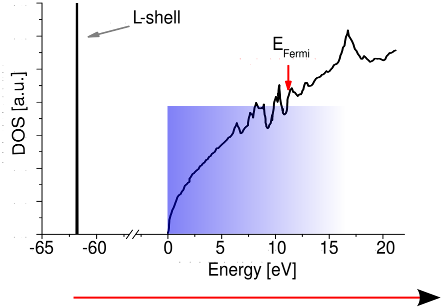
$$
\begin{aligned}
E_{\mathrm{EEG}} &= E_{\mathrm{gap}} + \langle E_e \rangle + \langle E_h \rangle \\
&= \frac{1}{2}\left(E_{\mathrm{gap}} + \min(E_e) + \min(E_h)\right)
\end{aligned}
$$



2. XUV irradiation

Excitation of aluminum

Metal: conduction band contains electrons



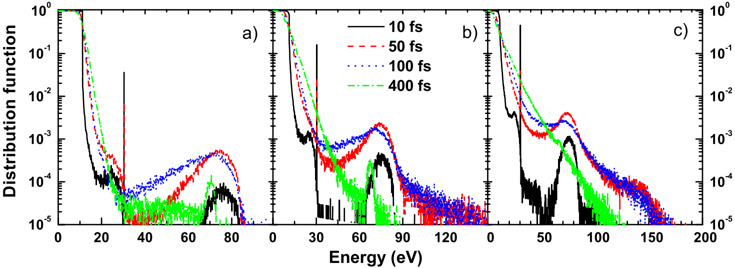
$\hbar\omega_L = 92$ eV

How do conduction band electrons and ionized electrons influence each other?

2. XUV excitation of aluminum

Transient distribution function

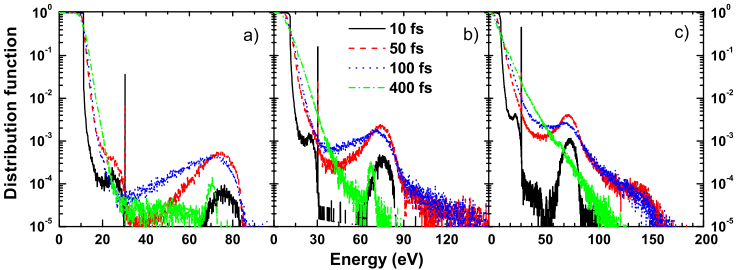
Increasing fluence ⟶



2. XUV excitation of aluminum

Transient distribution function

Increasing fluence ⟶



- Low–energy electrons close to Fermi distribution
- High–energy tail with Auger–bump survives > 100 fs
- Agrees with both experimental results:
  U. Zastrau et al., PRE **78**, 066406 (2008) & S. Vinko et al., PRL **104**, 225001 (2010)

Medvedev, Zastrau, Förster, Gericke, Rethfeld, PRL **107** 165003 (2011)

3. Hybrid simulation

Combination of
two-temperature description ...

$$\frac{\partial u_e}{\partial t} = -\alpha\,(T_e - T_{\mathrm{ph}})$$

$$\frac{\partial u_{\mathrm{ph}}}{\partial t} = +\alpha\,(T_e - T_{\mathrm{ph}})$$

3. Hybrid simulation

Combination of
two-temperature description ...

$$\frac{\partial u_e}{\partial t} = -\,\alpha\,(T_e - T_{\mathrm{ph}})$$

$$\frac{\partial u_{\mathrm{ph}}}{\partial t} = +\,\alpha\,(T_e - T_{\mathrm{ph}})$$

$$\Longrightarrow$$

... with Molecular Dynamics

$$\frac{\partial u_e}{\partial t} = -\,\alpha\,(T_e - T_{\mathrm{ph}})$$

$$m_i\,\frac{d^2\,\mathbf{r_i}}{dt^2} = \mathbf{F_i} + \zeta\,m_i\,\frac{d^2\,\mathbf{r_i^T}}{dt}$$

3. Hybrid simulation

Combination of two-temperature description ...

$$\frac{\partial u_e}{\partial t} = -\alpha\,(T_e - T_{\mathrm{ph}})$$

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Transport and excitation included

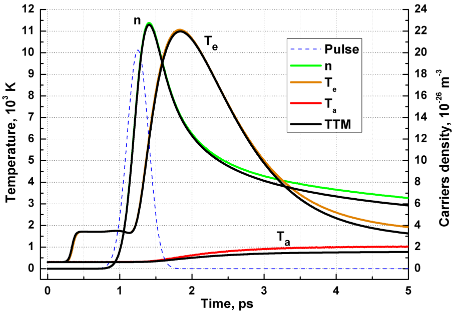
H. M. van Driel., Phys. Rev. B, **35** 8166-8176 (1987)

Energy conservation ensured

Ivanov and Zhigilei, Phys. Rev. B, **68** 064114 (2003)

3. Hybrid simulation

Example for visible-light excitation of semiconductors

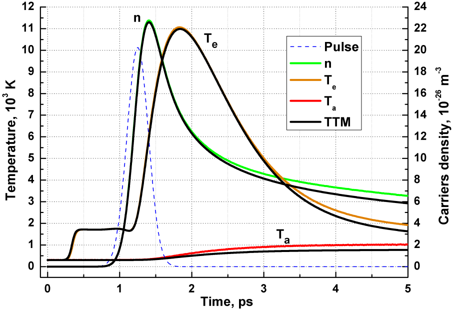


← nTTM (+ MD)

- Expansion changes temperature evolution

3. Hybrid simulation

Example for visible-light excitation of semiconductors



← nTTM (+ MD)

MD part →

- Expansion changes temperature evolution
- Amorphisation in the top 38 nm
- Close to experimental result
  J. Bonse, APA **84**, 63-66 (2006)

Summary

Calculate energy distribution

(methods: Boltzmann collision terms, Monte Carlo simulation)

↪ evolution of electron density

↪ energy transfer to lattice/atoms

1. Visible light
2. XUV irradiation
    - Water → to be combined with Molecular Dynamics
    - Silicon → Pair creation energy to estimate electron density
    - Aluminum → strong nonequilibrium, no single temperature
3. Hybrid simulation including lattice dynamics

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Thanks!

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