Theoretical approach to estimate radiation damage within FEL irradiated samples

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I. Mechanisms

Radiation damage

Contribution of different processes to radiation damage strongly depends on radiation wavelength

Radiation damage by VUV photons

Basic processes contributing:

- photoionizations (from outer shells) and collisional ionizations, elastic scatterings of electrons on atoms/ions
- long-range Coulomb interactions of charges with external and internal fields
- heating of electrons by inverse bremsstrahlung
- modification of atomic potentials by electron screening and ion environment
- recombination (3-body recombination)
- short range electron-electron interactions

Radiation damage by VUV photons

Also other processes may contribute:

- sequential ionization, e.g. $Xe \to Xe^*$, $Xe^* \to Xe^+$
- multiphoton ionization
- many-body recombination
- ionization by internal electric field (at the edge of the sample)
- ....

Radiation damage by VUV photons

Specific interactions in detail:

plasma effects

Plasma created by VUV photons

Example:

Xenon clusters $\rightarrow$ atomic density, $n_A = 10^{22} - 10^{23}$ cm^{-3} $\rightarrow$ estimated electron density $n_e = 10^{22} - 10^{24}$ cm^{-3} $\rightarrow$ dense plasma

Photoelectron energies, $E_{ph}(\lambda = 98\ nm) \approx 0.6$ eV
$E_{ph}(\lambda = 32\ nm) \approx 26.6$ eV

$\downarrow$

Temperature of emitted photoelectrons,

$T_{ph}(\lambda = 98\ nm) \approx 0.4$ eV $\leftarrow$ cold plasma
$T_{ph}(\lambda = 32\ nm) \approx 17.6$ eV $\leftarrow$ warm plasma

Plasma created by VUV photons

No experimental evidence on electron temperatures from first FEL experiment

At early stages:
cold, dense plasma
→ strongly coupled, degenerate plasma (quantum treatment of many body interactions necessary)

→ heats up

At later stages:
warm plasma → classical, ideal plasma (can be treated classically)

⇓

Difficult unified treatment of these two regimes

Plasma created by VUV photons

If heating mechanism efficient, classical treatment justified

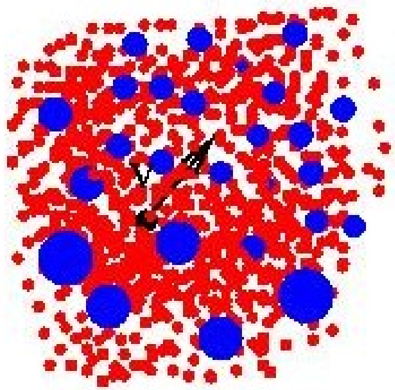
In any case we shall monitor Coulomb coupling and degeneracy parameters during the exposure ...

Plasma created by VUV photons

Cold, dense plasma effects

–degenerate: quantum statistics have to be used

–strongly coupled: electron and ions are quasiparticles with density dependent self-energies moving inside a dense interacting medium



Screening by electron-ion medium:
depends on impact velocity, $\mathbf{v}$

$$V_{eff}(\mathbf{k},\omega) = \frac{V_{coul}(\mathbf{k})\,\delta(\omega-\mathbf{kv})}{\epsilon_e(\mathbf{k},\mathbf{kv})+\epsilon_i(\mathbf{k},\mathbf{kv})-1}$$

Static, if $\mathbf{kv} \approx 0$

Dynamic, if $\omega_e >> \mathbf{kv} >> \omega_i$ ← only electrons,

if $\omega_e >> \omega_i >> \mathbf{kv}$ ← electrons and ions,

ω_e, ω_i are plasma frequencies for electrons and ions

Plasma created by VUV photons

Effects of strong coupling and degeneracy:

–self energy of quasiparticles: electron and ions

–lowering of continuum level

–lowering of ionization thresholds

–merging of bound states into continuum at high densities (Mott effect)

–dynamic screening

–changes in photo-, collisional, and recombination rates

–Pauli blocking (phase-space occupation effects)

Plasma created by VUV photons

Proper treatment of cold dense plasma possible with complicated quantum kinetic equations [Rostock group]

Some dense plasma effects can be included into our semi-classical model:

–quasiparticle self energies

–lowering of continuum level

–lowering of ionization thresholds

–changes in photo-, collisional, and recombination cross sections due to the screening

Radiation damage by VUV photons

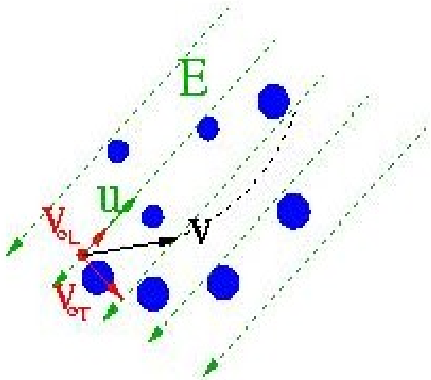
Specific interactions in detail:

inverse bremsstrahlung

Inverse bremsstrahlung

Classical impact model to start with:

elastic scatterings of electron on ions → gain of electron thermal energy due to momentum transfer



Initial and final electron velocities:
$\mathbf{v} = \mathbf{u} + \mathbf{v}_0$ and $\mathbf{v}' = \mathbf{u} + \mathbf{v}'_0$
$\mathbf{v}_0; \mathbf{v}'_0$, thermal velocities,
$\mathbf{u} = -\frac{e\,\mathbf{E}_0}{m\omega} sin(\omega t)$, quiver velocity,
where $E = E_0 \epsilon\, cos(\omega t)$, strength of electri
field of polarization ϵ

↓

Thermal energy gain, $\Delta\mathcal{E}_0 = -m\mathbf{u}(\mathbf{v}'_0 - \mathbf{v}_0)$

Inverse bremsstrahlung

Average energy gain per unit time

$$\left\langle \frac{d\mathcal{E}_0}{dt} \right\rangle_i = \frac{n_i i^2 e^4}{4\pi \epsilon_0^2} \left\langle \frac{\mathbf{u}\mathbf{v}}{v^3} \, ln\, \Lambda \right\rangle$$

where:

n_i, ion density in plasma

$ln\,\Lambda$, Coulomb logarithm dependent on cut-offs, $ln\,\Lambda \sim ln\,(b_{max}/b_{min})$, $b_{max} = v/\omega$, $b_{min} = max(\lambda_{Broglie}, b_{90^\circ})$

i, charge of a point-like ion

Inverse bremsstrahlung

Classical impact model cannot be applied in case of cold photoelectrons, as $b_{max} < b_{min}$

Inverse bremsstrahlung

Quantum emission or absorption of n photons [Kroll, Watson]:

$$\left(\frac{d\sigma}{d\Omega}\right)_{n,i}(\mathbf{v}_0; \mathbf{v}_0'; n\hbar\omega) \sim J_n{}^2\left(-\frac{e\,E_0}{m\omega}\frac{\epsilon\mathbf{Q}}{\hbar\omega}\right)\cdot \mid U(\mathbf{Q}) \mid^2,$$

where:

$\mathbf{Q} = m(\mathbf{v}_0' - \mathbf{v}_0)$, momentum transfer

$s = \frac{e\,E_0}{m\omega}\frac{1}{\hbar\omega}$, field strength parameter

$\mathcal{E}_0' = \mathcal{E}_0 + n\hbar\omega \; (\mid n \mid > 0\,)$, energy gain or loss

$U(\mathbf{Q})$, Fourier transform of the interaction potential, $U(\mathbf{r})$

Inverse bremstrahlung

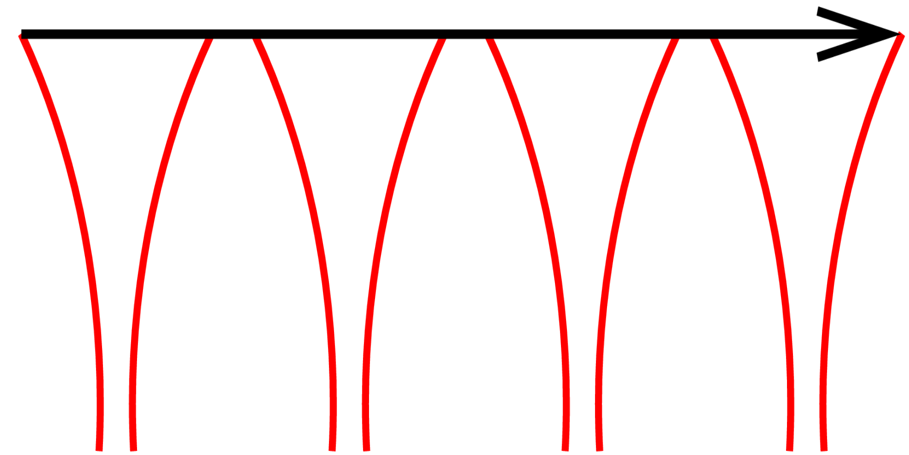
Field strength parameter, s:

$s << 1 \rightarrow J_n^2(sx) \sim (sx)^{2n} \rightarrow$ one-photon exchanges dominate

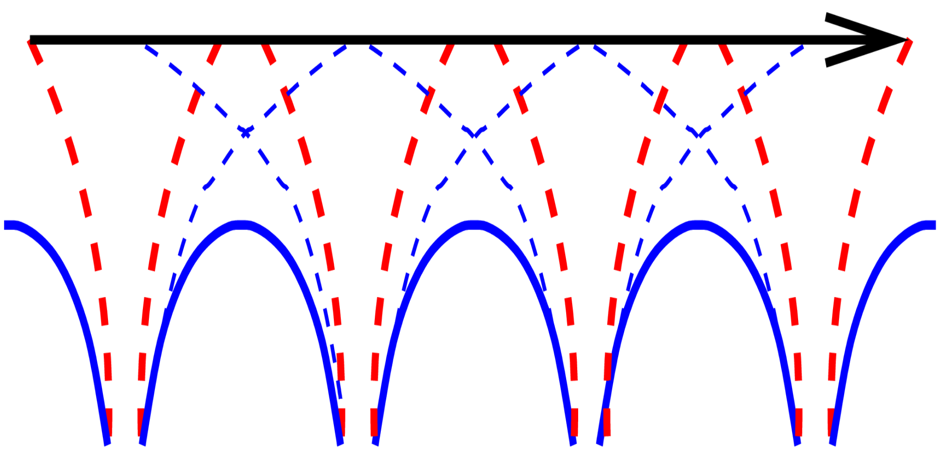
$s > 1 \rightarrow$ many-photon exchanges take place $\rightarrow$ limiting case can be identified with the classical impact picture

Electron screening and ion environment

Example: Sample of Xe atoms



← before ionization (neutral)



← after ionization, if electrons stayed inside (ion potential screened by electrons)

←after ionization, if some electrons escaped (ion potential barriers overlap)

Radiation damage by X-ray photons

Other processes than in VUV case give dominant contribution to radiation damage: inner and outer shell ionizations. No inverse bremsstrahlung.

Warm/hot plasma is created.

Radiation damage by X-ray photons

Basic processes contributing:

- photoionizations (from outer and inner shells with subsequent Auger decays) and collisional ionizations, elastic scatterings of electrons on atoms/ions
- long-range Coulomb interactions of charges with internal fields
- modification of atomic potentials by electron screening and ion environment
- recombination (3-body recombination)
- short range electron-electron interactions
- Compton scattering

No inverse bremsstrahlung at these radiation wavelengths!

Radiation damage by X-ray photons

Specific interactions in detail:

photo- and collisional ionizations

Photoionizations

Photoelectric effect: the dominant source of radiation damage (~90 % of interactions for light elements C, N, O, S)

5% of photoemissions: outer-shell event, single electron emitted

95% of events: inner-shell event, Auger effect

Auger effect

$\Downarrow$

K-hole (lifetime 1-10 fs)

$\Downarrow$

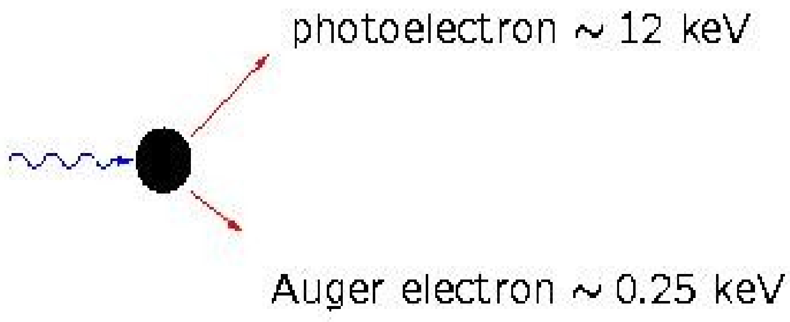
L-shell electrons fall into K-hole

$\Downarrow$

energy transfer

$\Downarrow$

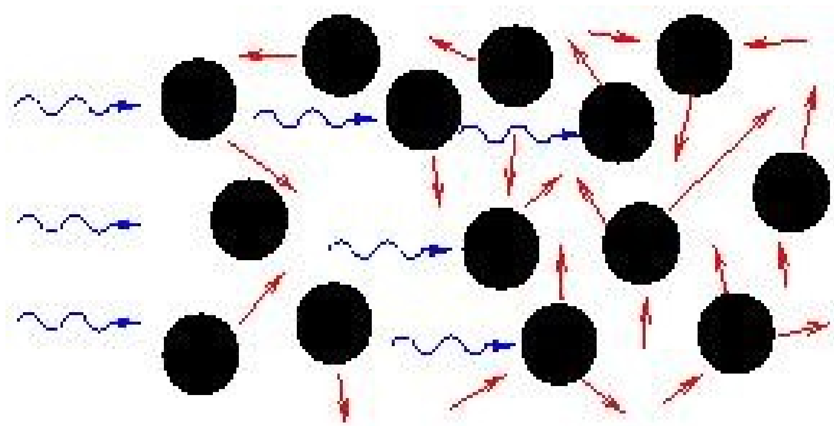
Auger electron $E \sim 0.25 keV$



Collisional ionizations by electrons

Primary ionization
(photo- and Auger)

Secondary ionization



Photons

Electrons

Collisional ionization by electrons

Two energy regimes of electrons released by X-ray photons:

- Photoelectrons: $E = 12$ keV, $\lambda_{DeBroglie} \approx$ 0.1 Å, fast, propagate almost freely through the medium, leave small samples (10-100 nm) in a few femtoseconds

- Auger electrons: $E = 0.25$ keV, $\lambda_{DeBroglie} \approx$ 0.8 Å, slow, interact multiply with neighbouring atoms, interaction includes exchange terms

Auger electrons are the main source of secondary ionization

II. Models

How to model radiation damage within irradiated samples?

Computer simulations of damage processes:

→ testing influence of specific interactions on ionization dynamics

→ accurate time characteristics of damage processes

Particle approach

Solving equations of motion for each particle at each time step

$$\frac{d\mathbf{r}}{dt} = \mathbf{v}$$

$$\frac{d\mathbf{v}}{dt} = \mathbf{a}$$

Results are averaged over the total numer of simulated events

Scattering probabilites: obtained with cross sections

Particle approach

Advantages:

- first-principles model
- transparent algorithm
- no complex numerics

Disadvantages:

- High computational costs which scale with the number of participating particles
- statistical errors

Particle approach

Methods:

- stochastic Monte-Carlo method
- deterministic Molecular Dynamics simulations
- Particle-in-Cell method

Particle approach

Example: MC code for modelling Auger-electron cascade in diamond

Electrons interact multiply with atomic clusters:

- elastically - with no energy loss

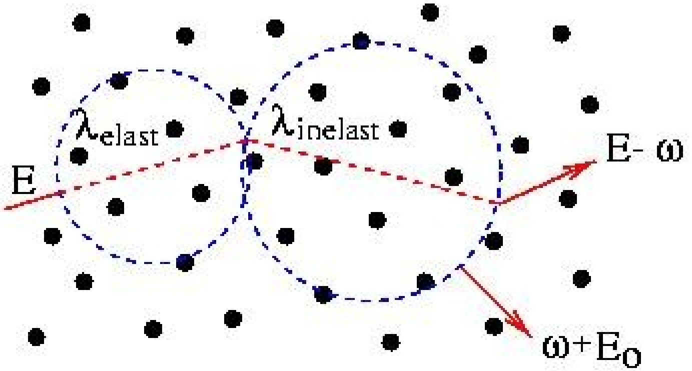
$$\lambda_{elast} \sim \frac{1}{\sigma_{elast}}$$

- inelastically - with energy loss ω, either transferred to a cluster, or to another electron

$$\lambda_{inelast} \sim \frac{1}{\sigma_{inelast}}$$

$$\Downarrow$$

Cascade of secondary electrons



Particle approach

Example: MC code for modelling Auger-electron cascade in diamond

- Elastic collisions: muffin-tin potential, partial wave expansion, phase shifts δ_l

$$\sigma_{elast} = \frac{4\pi}{k^2} \sum_{l=0}^{\infty} (2l+1) \sin^2 \delta_l$$

$$\Downarrow$$

$$\lambda_{elast} \sim \frac{1}{\sigma_{elast}}$$

- Inelastic collisions: optical models based on an atomic-oscillator model of dispersive media, dielectric function $\epsilon(q,\omega)$, differential inverse mean free path $\tau(E,\omega)$

$$\tau(E,\omega) \sim \int \frac{dq}{q} Im[-\epsilon(q,\omega)^{-1}]$$

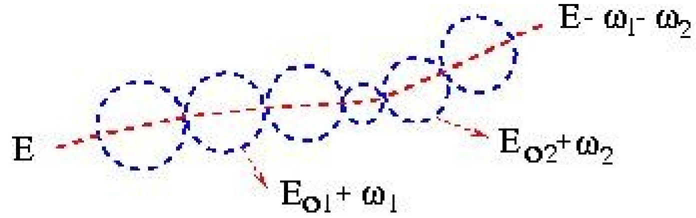
$$\Downarrow$$

$$\lambda^{-1}_{inelast} = \int d\omega \, \tau(E,\omega)$$

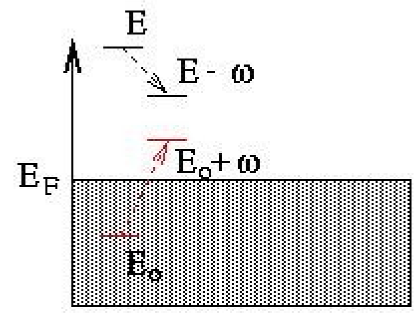
Particle approach

Example: MC code for modelling Auger-electron cascade in diamond

Time evolution of the cascade



E_{0i} depends on the electronic band structure.
Here: Fermi free-electron band.

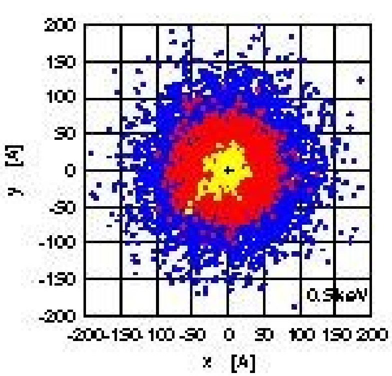


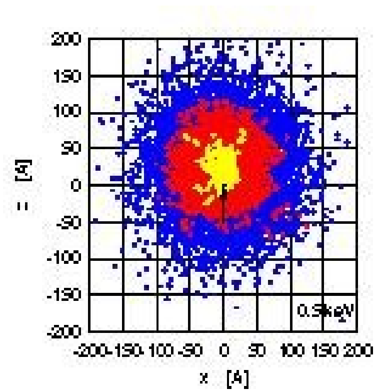
Particle approach

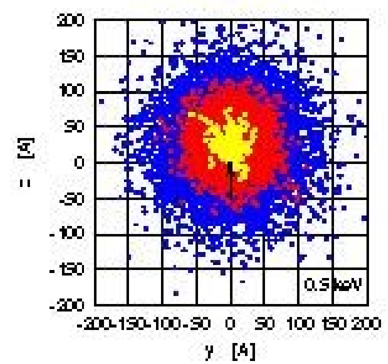
Example: MC code for modelling Auger-electron cascade in diamond

Results: spatio-temporal evolution of electron cascade

Electron range:



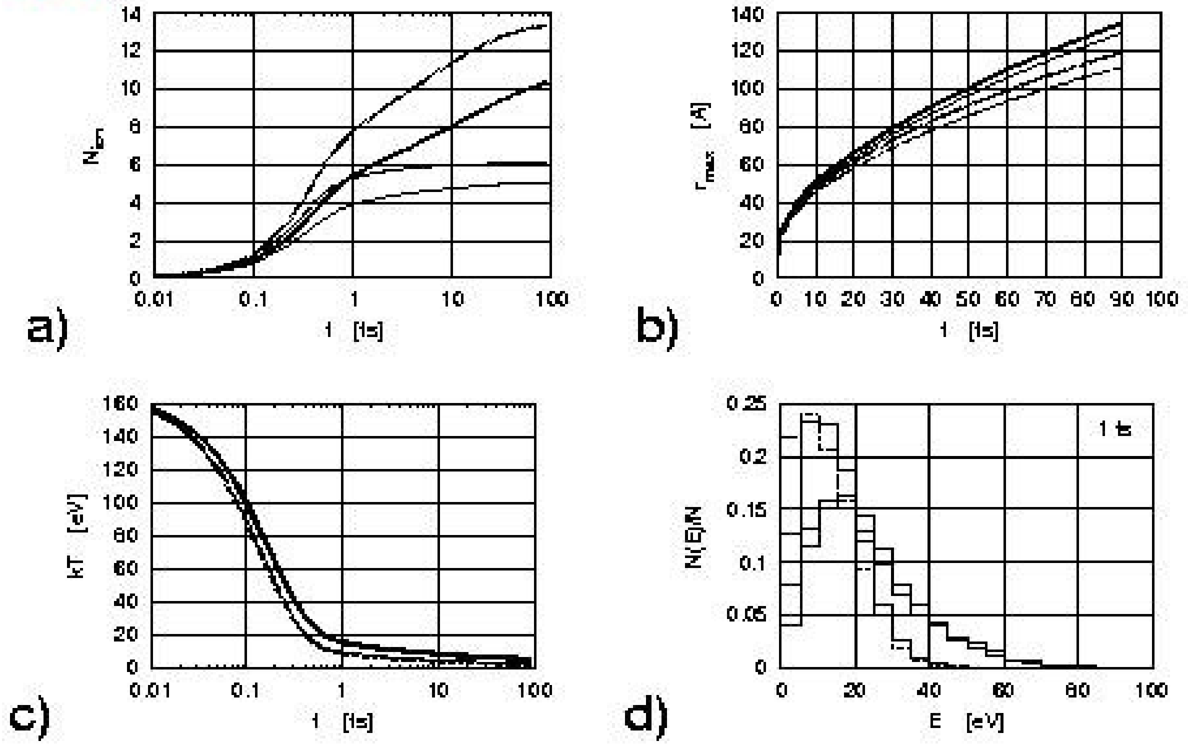




Particle approach

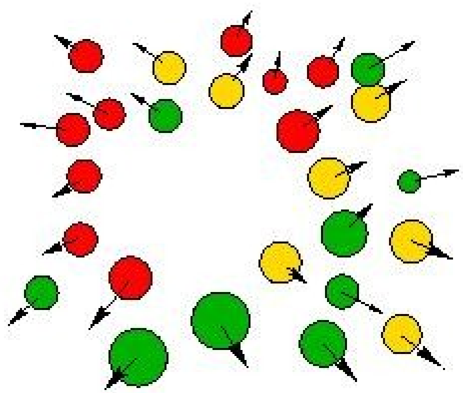
Example: MC code for modelling an Auger-electron cascade in diamond

Results: spatio-temporal characteristics of the cascade

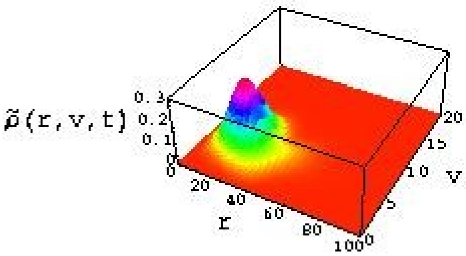


Transport approach

Evolution of larger systems described in terms of collective density function:



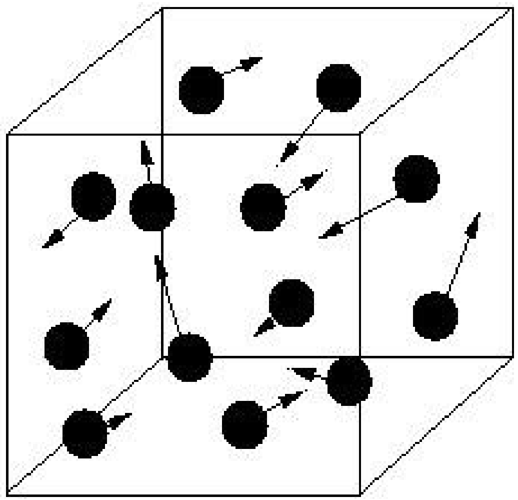
$\Longleftrightarrow$



Transport approach

Statistical description of a classical system in terms of density functions $\rho(\mathbf{r}, \mathbf{v}, t)$ in phase space

$\rho(\mathbf{r}, \mathbf{v}, t) d^3r\, d^3v$ is a number of particles located at $\mathbf{r}$ of velocity $\mathbf{v}$ in the phase space element $d^3r\, d^3v$



$$\int \rho(\mathbf{r}, \mathbf{v}, t) d^3r\, d^3v = N(t)$$

$$\int \rho(\mathbf{r}, \mathbf{v}, t)\; d^3r = n(\mathbf{v}, t)$$

$$\int \rho(\mathbf{r}, \mathbf{v}, t)\; d^3v = n(\mathbf{r}, t)$$

Transport approach

Methods:

- Semiclassical Boltzmann equation
- Hydrodynamic models

Transport approach

Time evolution of the system

$$t \rightarrow t + dt$$

$$\mathbf{r} \rightarrow \mathbf{r} + \mathbf{v}dt$$

$$\mathbf{v} \rightarrow \mathbf{v} + \mathbf{a}dt$$

$$\rho(\mathbf{r} + \mathbf{v}dt, \mathbf{v} + \mathbf{a}dt, t + dt)d^3r\, d^3v - \rho(\mathbf{r}, \mathbf{v}, t)d^3r\, d^3v = 0$$

Boltzmann equation

$$\partial_t \rho + \mathbf{v}\partial_{\mathbf{r}}\rho + \mathbf{a}\partial_{\mathbf{v}}\rho = 0$$

If there is a collision or a change of particle number:

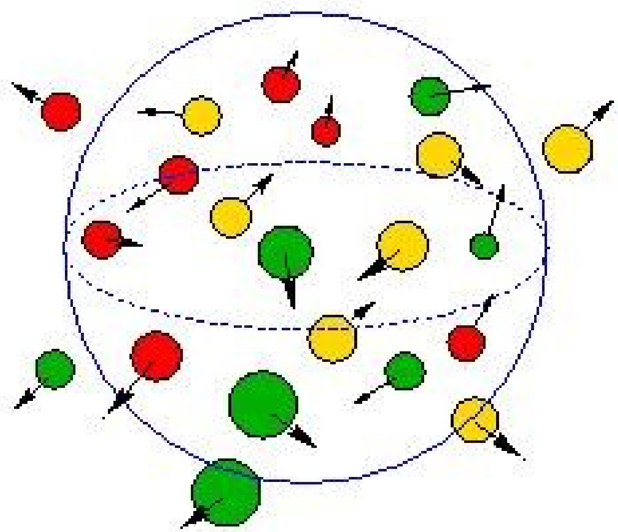
$$\partial_t \rho + \mathbf{v}\partial_{\mathbf{r}}\rho + \mathbf{a}\partial_{\mathbf{v}}\rho = \Omega(\rho, \mathbf{r}, \mathbf{v}, t),$$

where Ω is a source (collision) term.

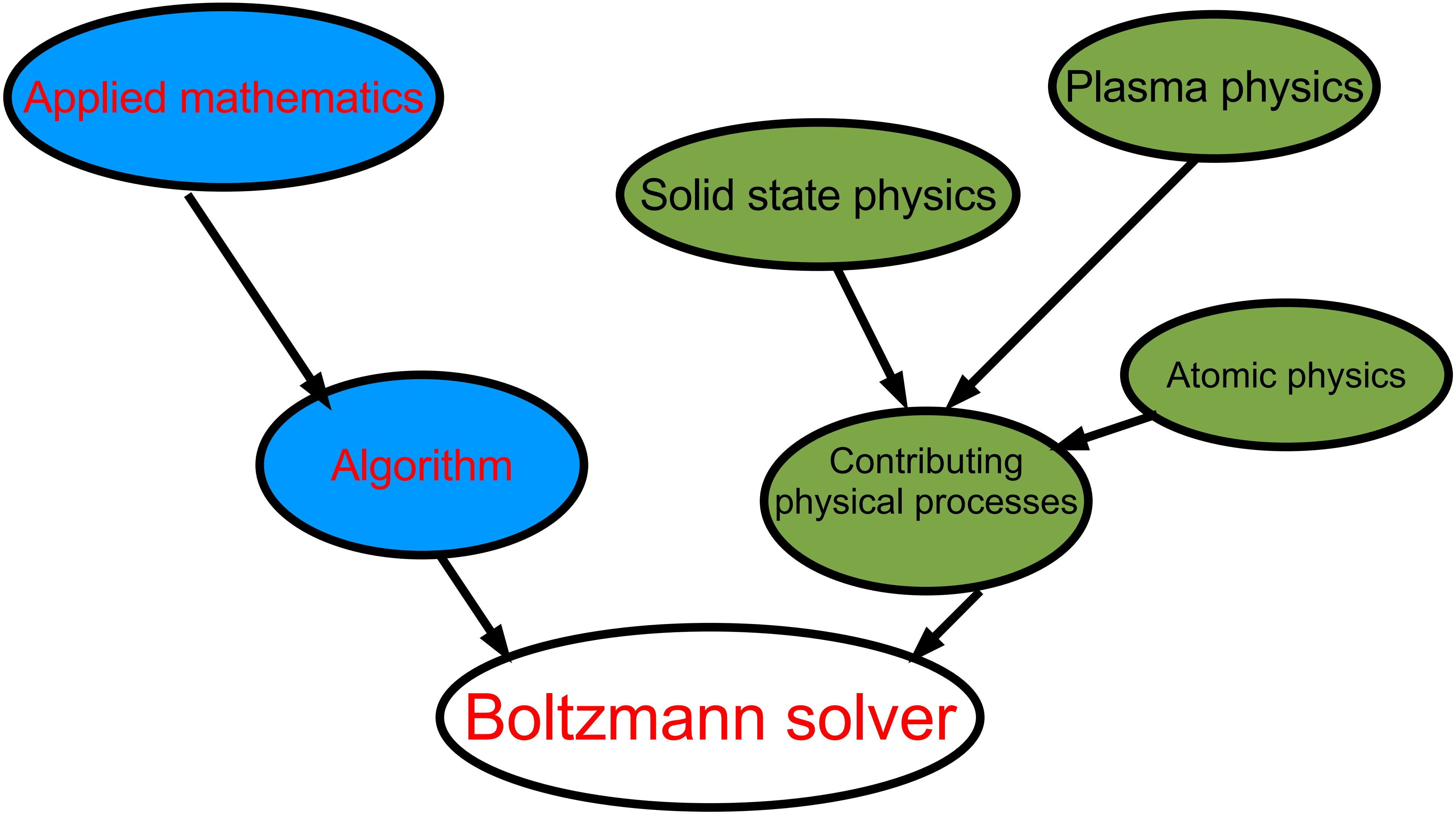
Transport approach

Boltzmann equations are able to follow

non-equilibrium processes



Working with Boltzmann equations



Why to use statistical Boltzmann approach?

- first-principle approach
- single-run method
- computational costs do not scale with number of atoms

Disadvantage:

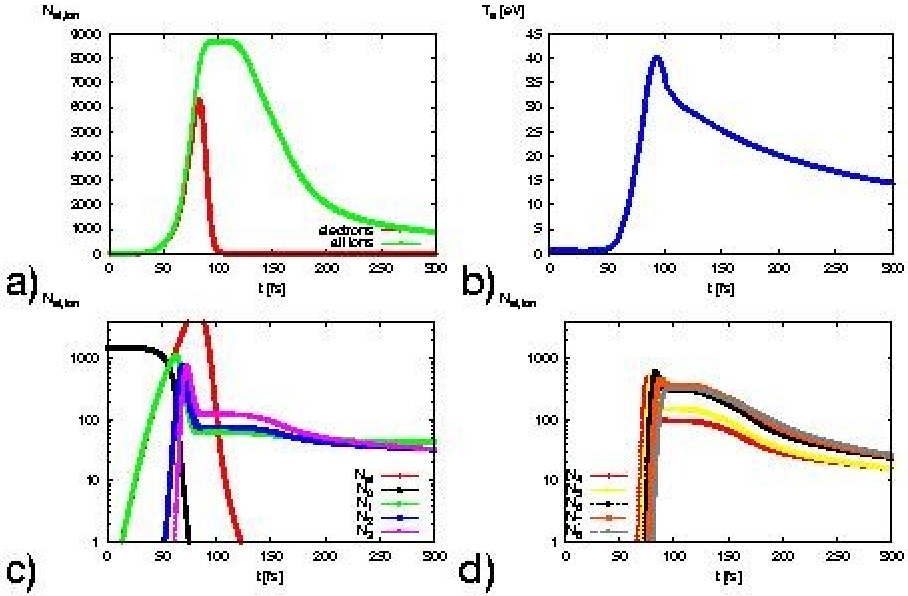
- requires advanced numerical methods

Transport approach

Example: Boltzmann solver for radiation damage within Xe clusters irradiated by VUV FEL photons

Global parameters as functions of time

Fast heating rate (with modified atomic potential)



a) total number of particles in the cluster, b) electron temperature, c) number of electrons and ions up to +3, and d) ions +4 up to +8

Hydrodynamic models

Reduced form of Boltzmann equations: simplifying assumptions applied (e.g. only collective transport component of velocity treated, thermalisation of electrons assumed, local force equilibrium assumed) - standard tool for plasma simulations

Conclusions, questions and outlook

- Understanding of radiation damage within FEL irradiated samples important for theory and experiment

- Description of radiation damage more complicated in case of irradiation with VUV photons that with X-ray photons

- Choice of the method for modelling the radiation damage depends on the size and structure of the sample:

  (particle method for smaller, complex samples;

  transport method for large samples of regular structure)