MAMBA
Materials irradiation
from basics to applications

Combining transport Monte Carlo with other simulation methods

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Outline

1. Introduction: what is MC and applications
2. Example: MC integration
3. Random number generators
4. MC in radiation transport
5. Cross sections
6. Photons, electrons, holes, ions
7. Combining MC with other methods: hybrid, multiscale…
8. Models without feedbacks
9. Models with feedbacks

Introduction: Monte Carlo

N. Metropolis, S. Ulam
"The Monte Carlo Method“
J. Amer. Stat. Assoc. 44, 335 (1949)

Key to Monte Carlo method: randomness

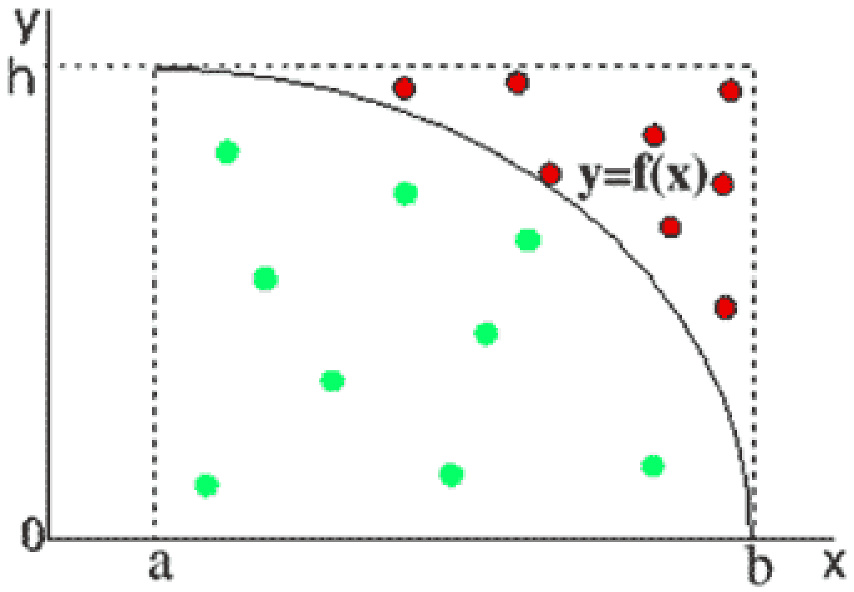
Monte Carlo methods (plural)

There is no “**the”** Monte Carlo (MC) method: huge variety of them

- Metropolis MC (ensemble evolution)
- Biology MC
- Comparison of risk analysis (investment banking)
- Direct simulation MC (gas and fluid flow)
- Dynamic MC (chemistry)
- Kinetic MC (defects in solids)
- Quantum MC
- **MC for particle transport**
- **Event-by-event (analog) MC**
- Condensed collisions MC
- etc.

Example: Monte Carlo integration

Solving integrals with MC method (a.k.a. rejection method)



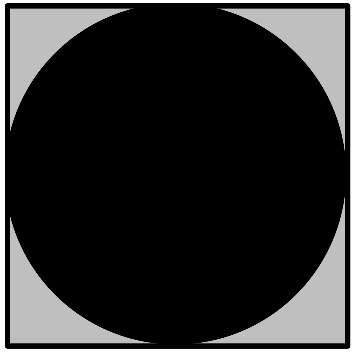
Area under a curve is ~ number of points under it vs. total

$$\int_a^b f(x)\, dx \approx S_0 \frac{N_{in}}{N}$$

Convergence

Number of iterations defines variance: $\sigma_Q = \sqrt{\frac{\mathrm{var}(q)}{N}} = \sqrt{\frac{1}{N}\left[\frac{1}{N}\sum_{i=1}^{N} q_i^2 - \overline{Q}^2\right]}.$

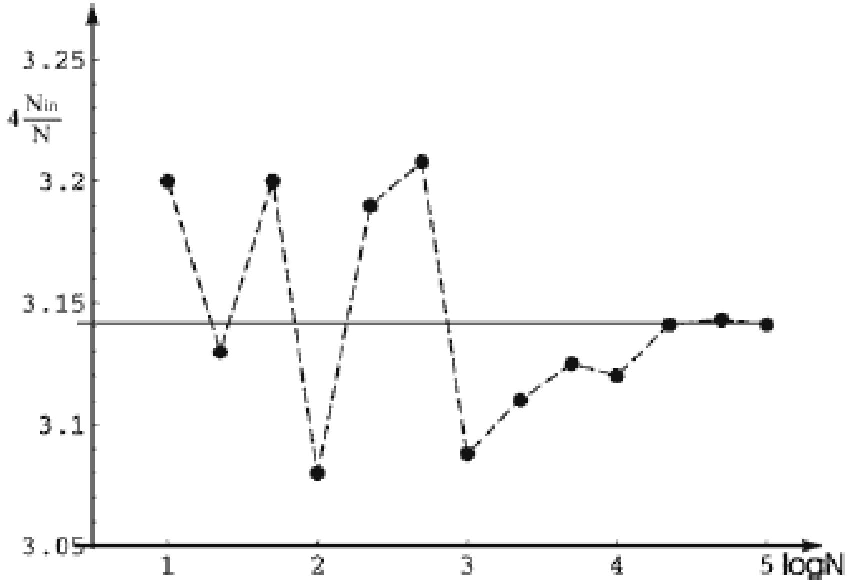
Example: circle in a square



$$x^2 + y^2 < R^2$$

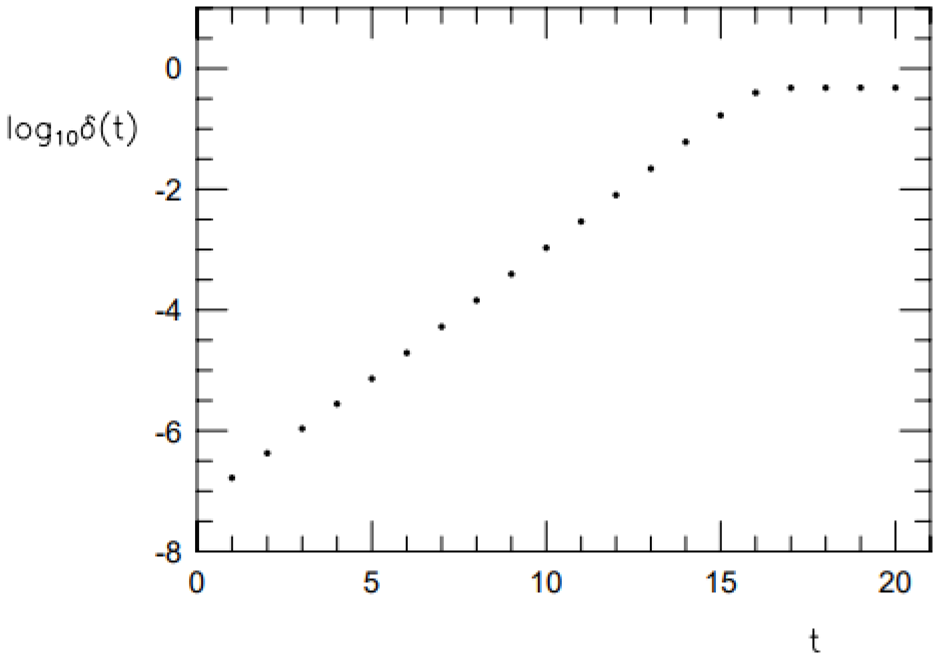
$$S_{in} = S\,\frac{N_{in}}{N}$$

$$\pi = 4\frac{N_{in}}{N}$$



Increasing number of iterations decreases variance

Random number generators



Theoretically proven randomness:

ranlux: http://luscher.web.cern.ch/luscher/ranlux/

mixmax: https://mixmax.hepforge.org/

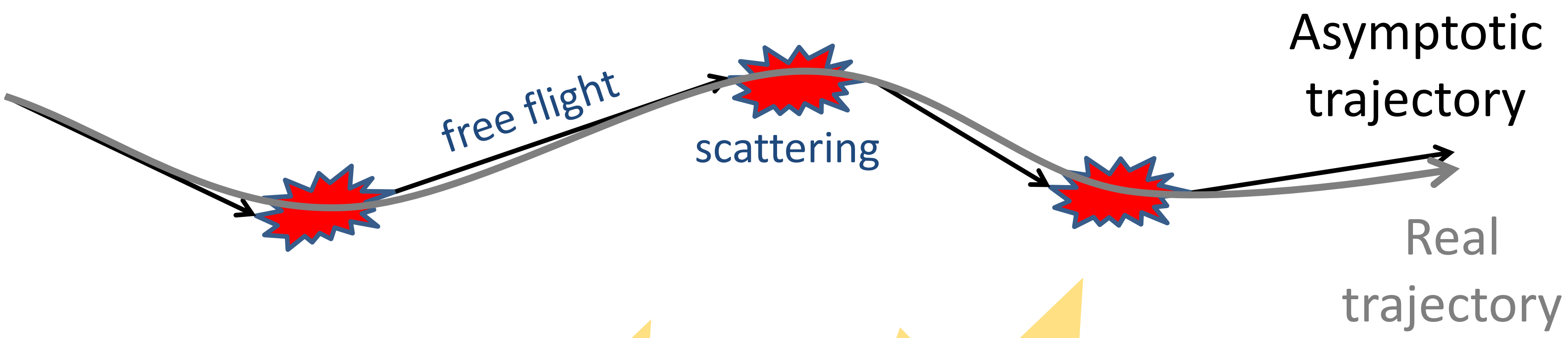
Both are available for FORTRAN and C

Radiation transport

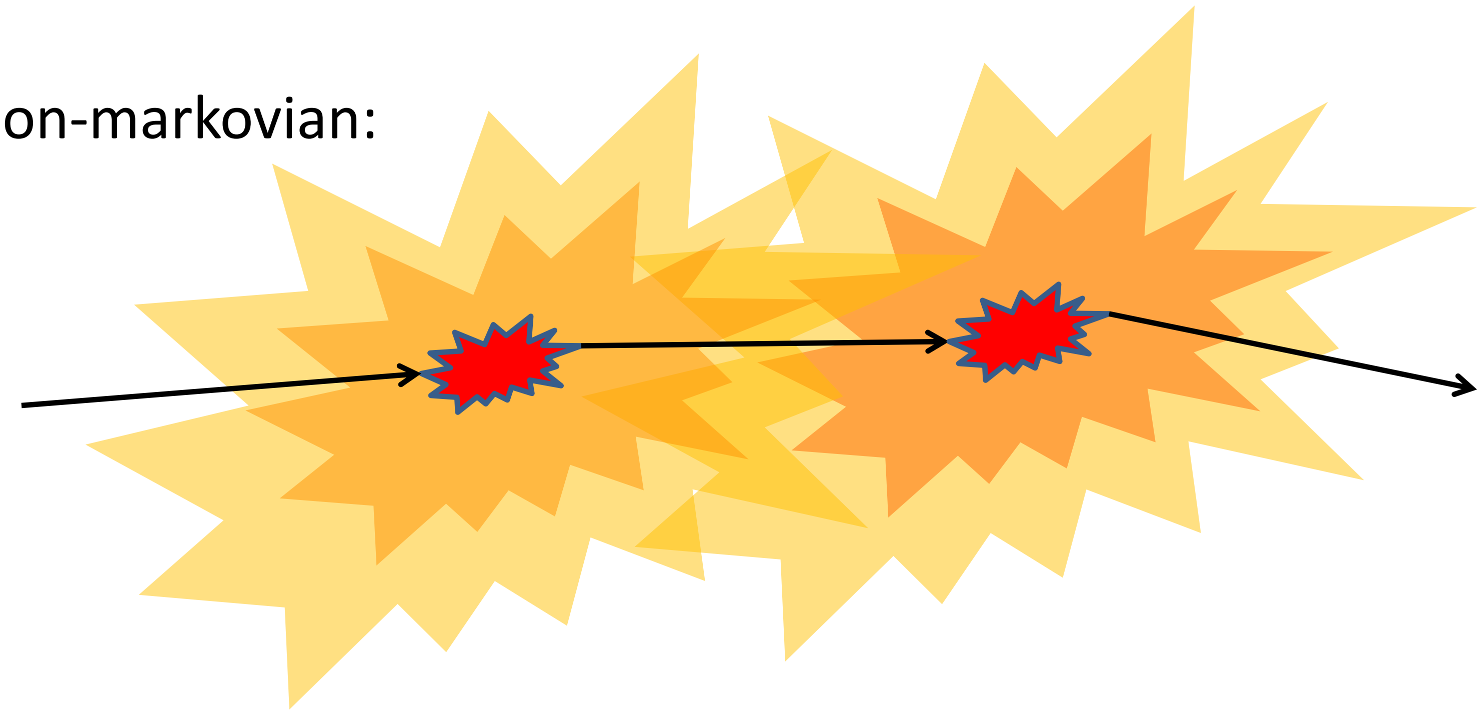
“Tools for investigating electronic excitation: experiment and multi-scale modelling”
Instituto de Fusión Nuclear "Guillermo Velarde", Universidad Politécnica de Madrid
https://doi.org/10.20868/UPM.book.69109

Radiation transport

Markov chain: duration of scattering event << free flight



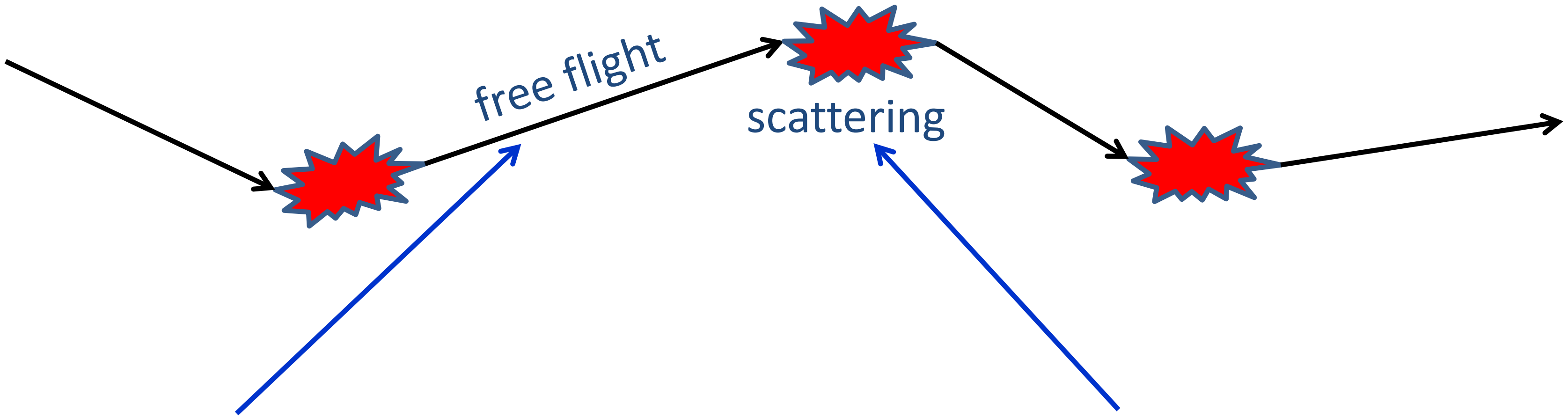
Non-markovian:



Keep in mind quantum speed limit theorem!

Monte Carlo of radiation transport

Quantities defined by probabilities:



Free flight distance

$$l = -\lambda \ln(\gamma)$$

Mean free path: $\lambda = \dfrac{1}{n_e \sigma}.$

Scattering event:

- What kind of event?
- How much energy is lost?
- How is momentum changed?

“Tools for investigating electronic excitation: experiment and multi-scale modelling”
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Monte Carlo: key values

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Sampled flight distance: $l = -\lambda \ln(\gamma)$ $\lambda = \frac{1}{n_e \sigma}$.

Mean free path: $\lambda^{-1} = n_{at} \int_{E_{min}}^{E_{max}} \int_{q_-}^{q_+} \frac{d^2\sigma}{d(\hbar\omega)d(\hbar q)} d(\hbar\omega)d(\hbar q)$

Stopping power: $S_e = -\frac{dE}{dx} = n_{at} \int_{E_{min}}^{E_{max}} \int_{q_-}^{q_+} \hbar\omega \frac{d^2\sigma}{d(\hbar\omega)d(\hbar q)} d(\hbar\omega)d(\hbar q)$

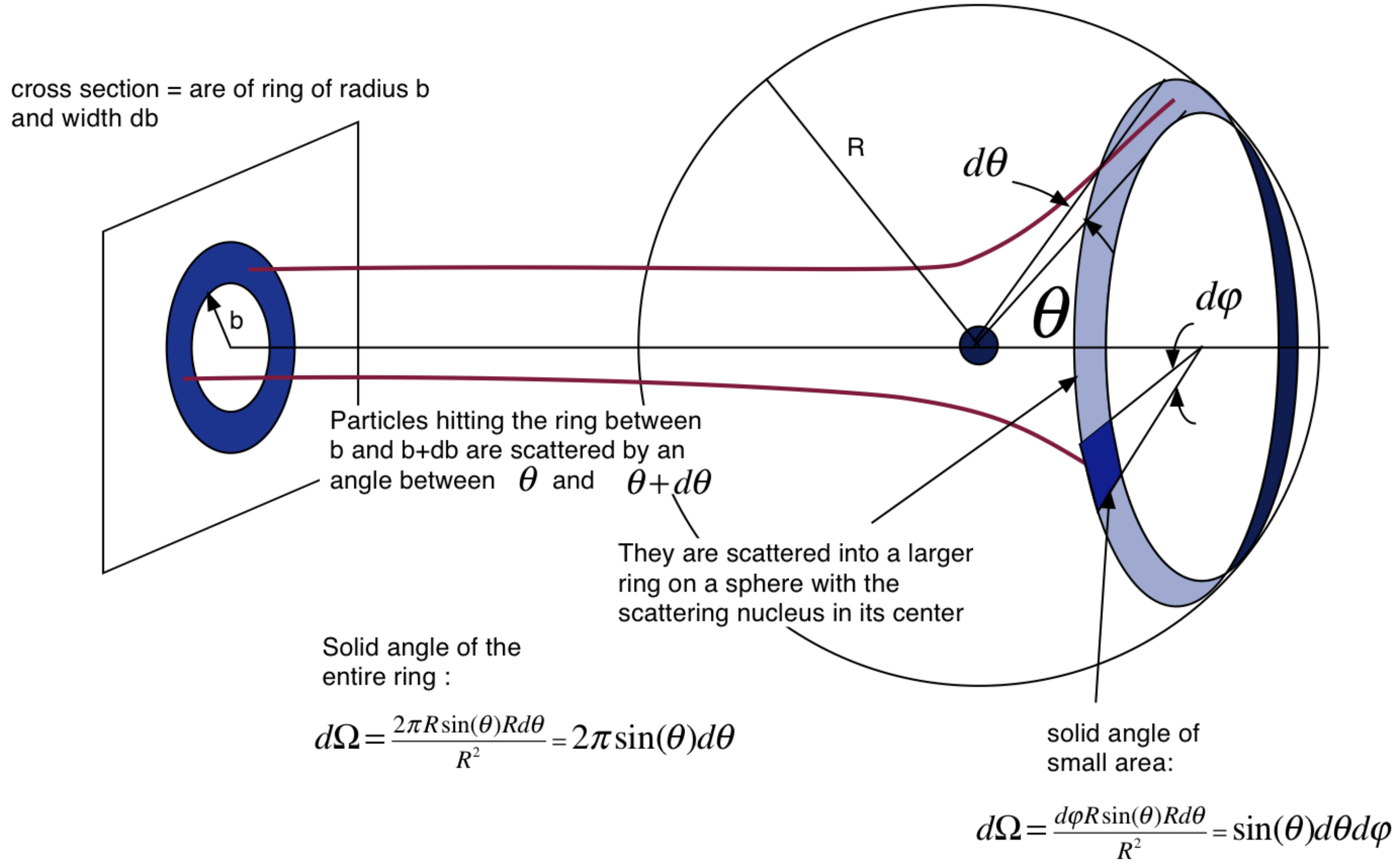
Kind of collisions: $P_i = \frac{\sigma_i}{\sum \sigma_i}$

Energy loss in a scattering event: $\gamma\sigma = \int_{E_{min}}^{\delta E} \int_{q_-}^{q_+} \frac{d^2\sigma}{d(\hbar\omega)d(\hbar q)} d(\hbar\omega)d(\hbar q)$

In summary: cross sections define everything!

Cross sections

Cross sections



Area, where particles should meet to scatter

(for given energy, into given angle)

Pic from:
http://hep.physics.wayne.edu/~harr/courses/5210/w15/lecture29.htm

Probability of scattering ~ cross section

Cross sections

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L. Van Hove, Phys. Rev. **95**, 249 (1954)

Plane waves: $|k^e_{i,f}\rangle = V^{-\frac{1}{2}}\exp(i\boldsymbol{k}^e_{i,f}\,\boldsymbol{r})$ <- Fourier transform

Dynamic structure factor:

$$S_{ab}(q,\omega) = \sum_i P_i \left\langle i \right| \int \frac{dt}{2\pi} \exp(i\omega t) \int d\boldsymbol{r} \int d\boldsymbol{r}' \exp\left(-i\boldsymbol{q}(\boldsymbol{r}-\boldsymbol{r}')\right) \hat{n}_a(\boldsymbol{r},t)\hat{n}_b(\boldsymbol{r}',0) \left| i \right\rangle$$

First Born approximation: $$\frac{d^2\sigma_{e-at}}{d(\hbar q)d(\hbar\omega)} = \frac{q}{2\pi\hbar^4}\frac{1}{v^2}\left(\frac{4\pi e^2}{q^2}\right)^2 (Z)^2 S_{ii}(\omega,q)$$

Scattering on individual atom

Collective behavior of the system

Cross sections

Fluctuation dissipation theorem:

$$Im\left[-\frac{1}{\varepsilon(\omega,q)}\right]=\frac{4\pi e^2}{q^2\hbar}\left(1-e^{-\frac{\hbar\omega}{T}}\right)S(\omega,q)$$

$$\frac{d^2\sigma}{d(\hbar\omega)d(\hbar q)}=\frac{2\left(Z_{eff}(v)e\right)^2}{n_{at}\pi\hbar^2v^2}\frac{1}{\hbar q}\left[1-\exp\left(-\frac{\hbar\omega}{k_BT}\right)\right]^{-1}Im\left(\frac{-1}{\varepsilon(\omega,q)}\right)$$

Individual atom — **Thermal factor** — **Loss function**

“Tools for investigating electronic excitation: experiment and multi-scale modelling”
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Photons

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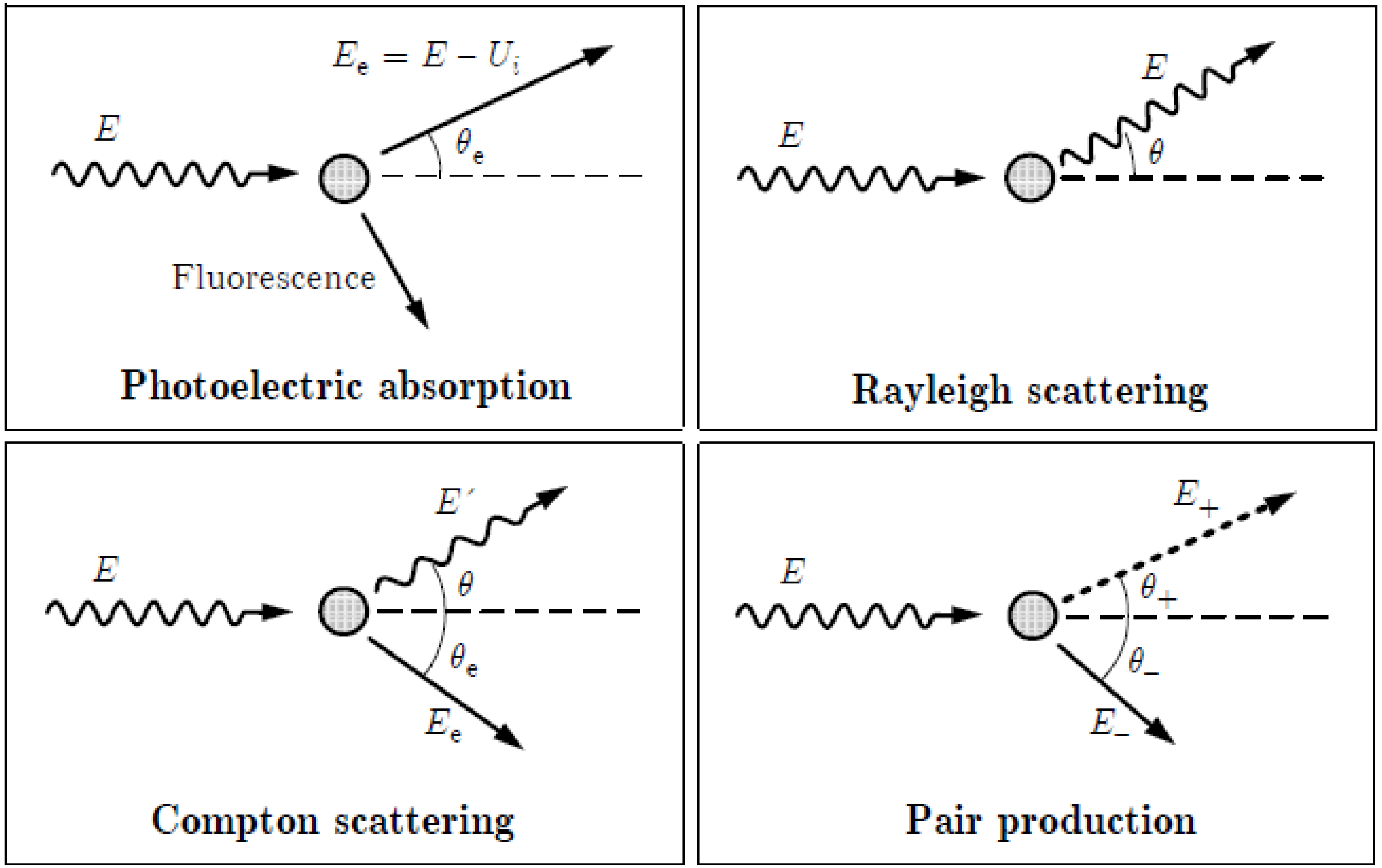
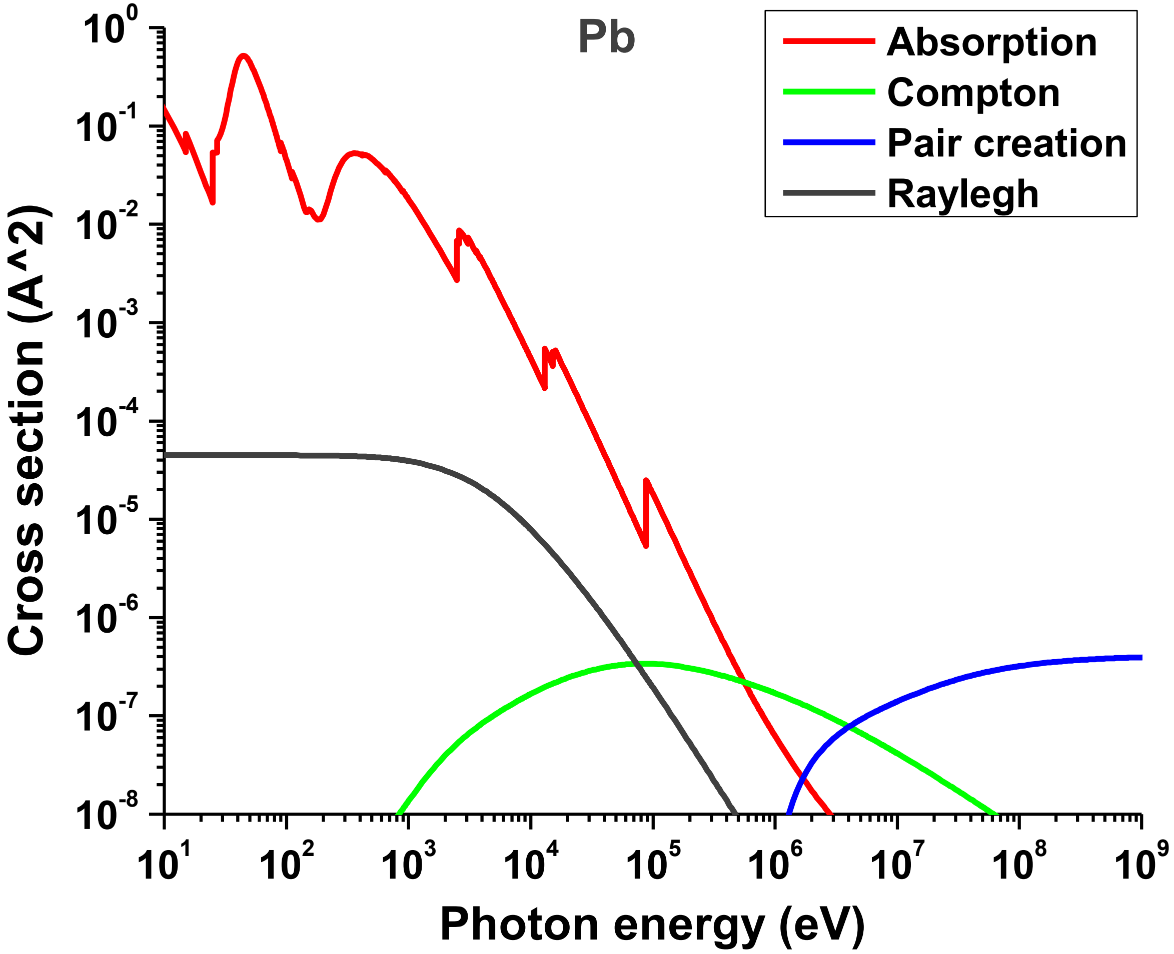
Photons

Scattering processes:



F. Salvat, J.M. Fernzndez-Varea, J. Sempau “PENELOPE 2014 - A Code System for Monte Carlo Simulation of Electron and Photon Transport” (2014)

Photon cross sections



EPICS2023 database: https://www-nds.iaea.org/epics/

Henke's tables: http://henke.lbl.gov/optical_constants/atten2.html

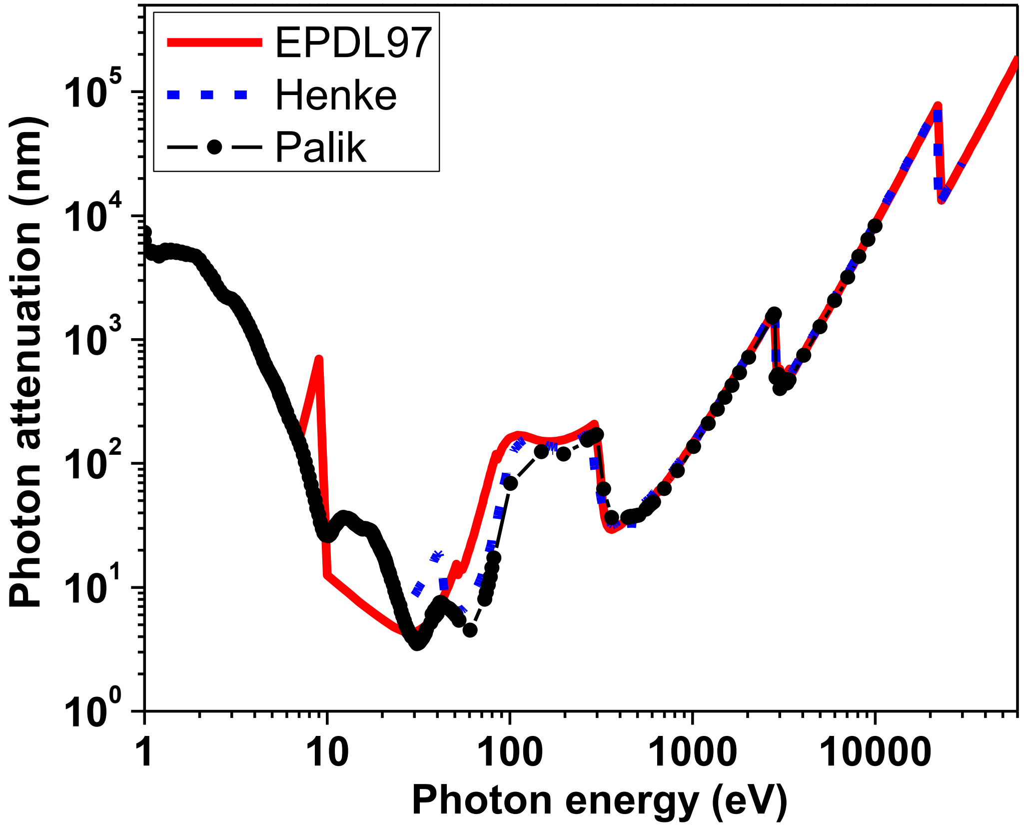
Photoabsorption: collective effects

In solids – band effects at low energies

Experimental optical coefficients:

$$\mathrm{Im}\left[\frac{-1}{\varepsilon(\omega, q=0)}\right] = \frac{2nk}{(n^2-k^2)^2+(2nk)^2}.$$

$$\mathrm{Im}\left[\frac{-1}{\varepsilon(\omega, q=0)}\right] = \frac{c}{\lambda_{ph}\omega},$$

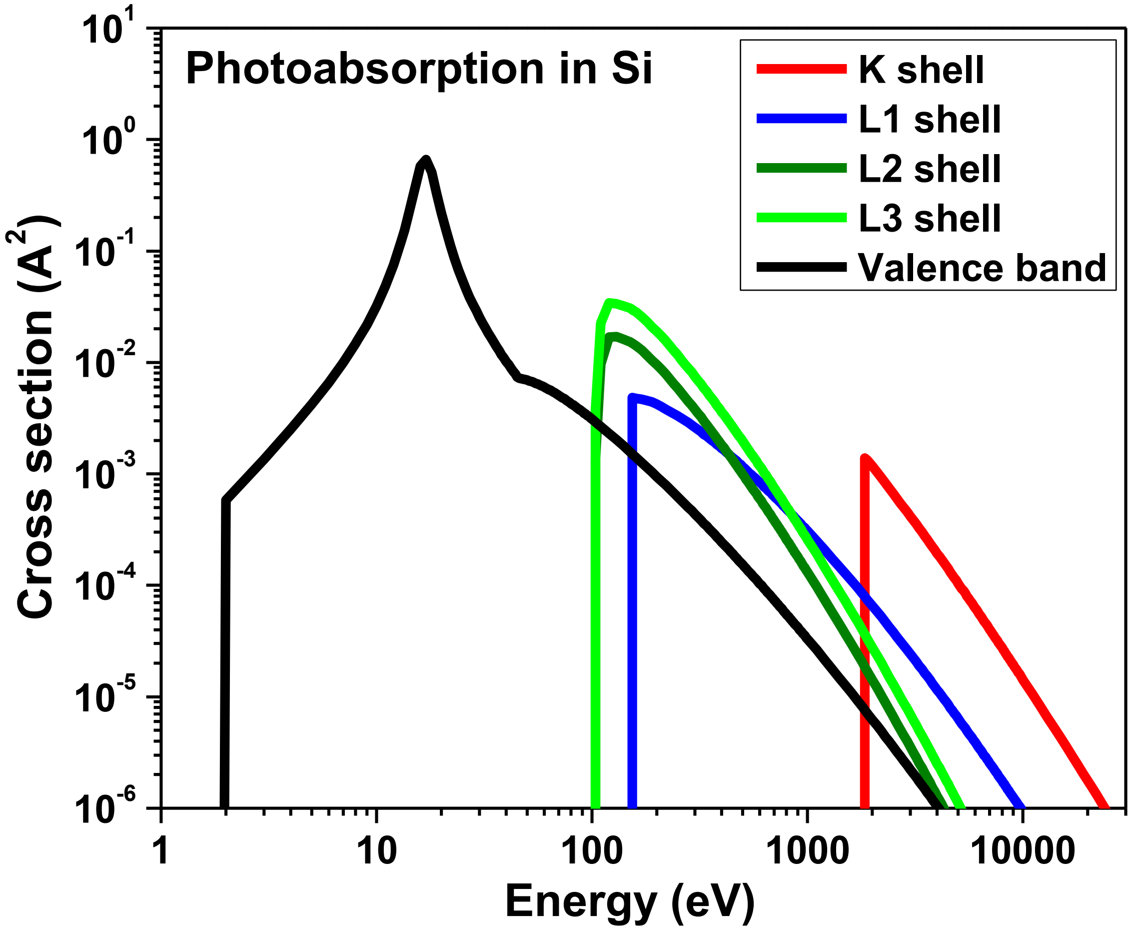


Milov *et al.*, JOSA B 35, B43 (2018)

For photon energies below ~100 eV atomic cross sections differ from solids

Photoabsorption: collective effects

Atomic shell choice: $P_i = \frac{\sigma_i}{\sum \sigma_i}$



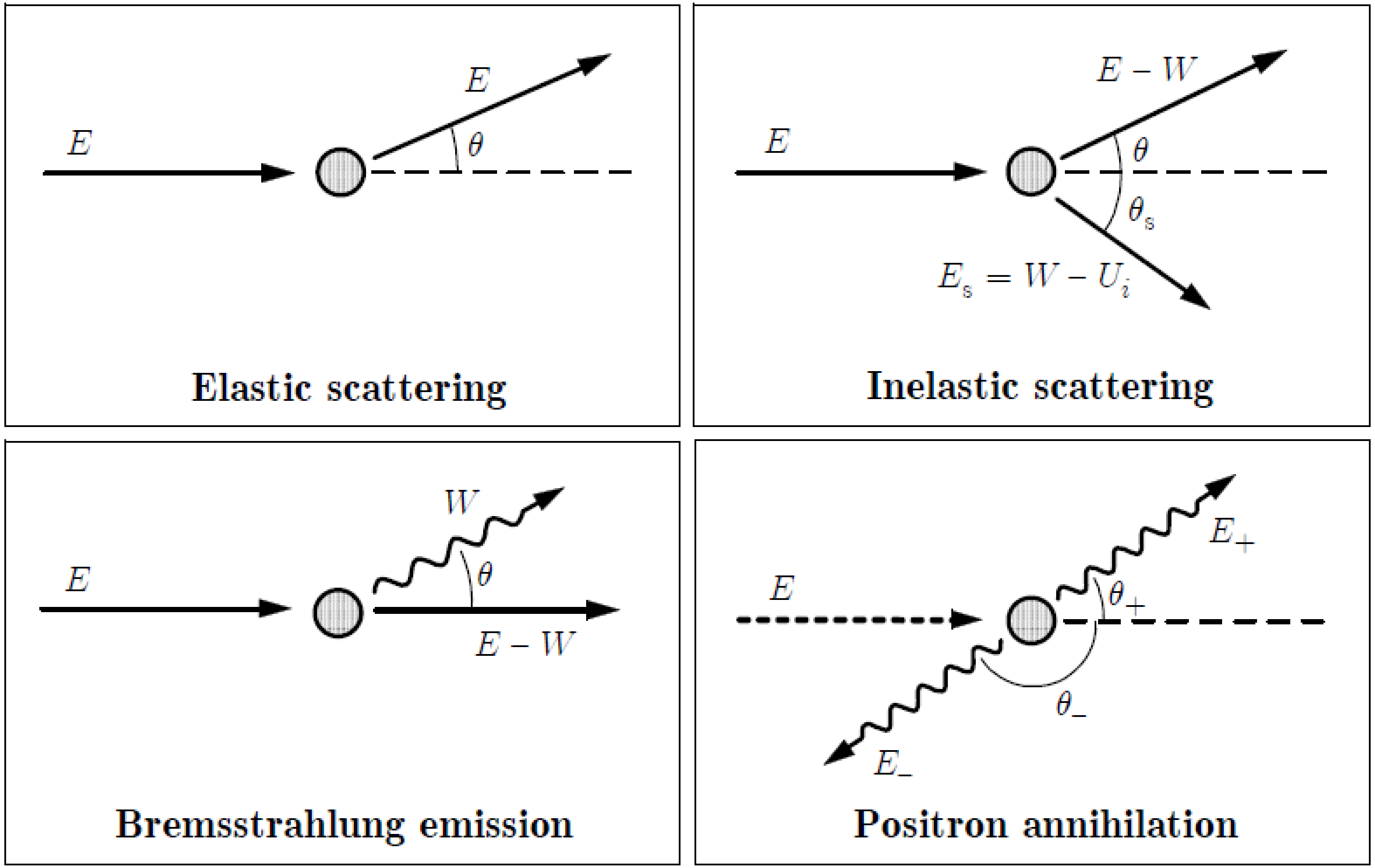
Photons: deepest shell ionization is most probable

[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

Electrons / ions / charged particles

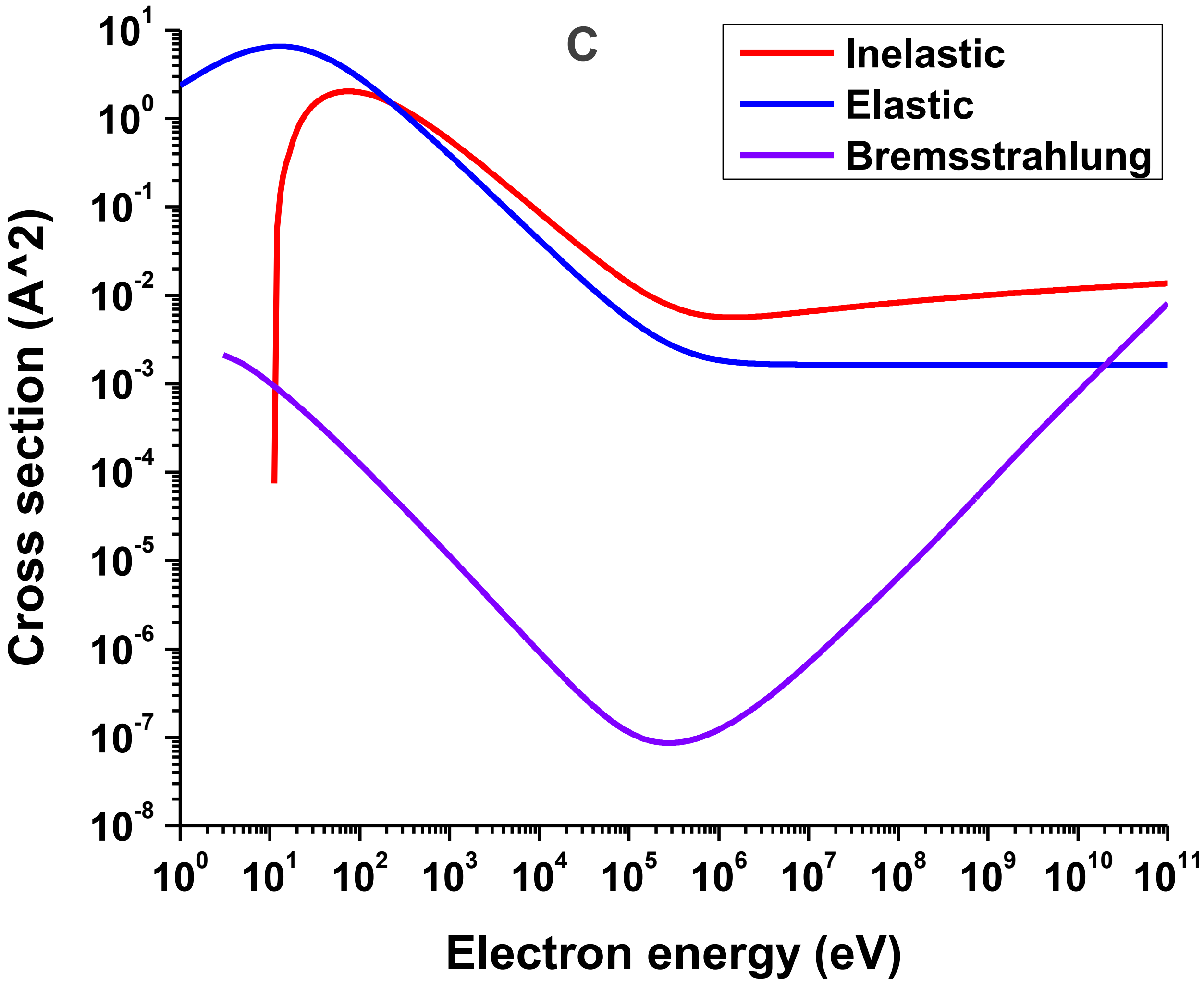
Electrons

Processes



[F. Salvat, J.M. Fernzndez-Varea, J. Sempau "PENELOPE 2014 - A Code System for Monte Carlo Simulation of Electron and Photon Transport" (2014)]

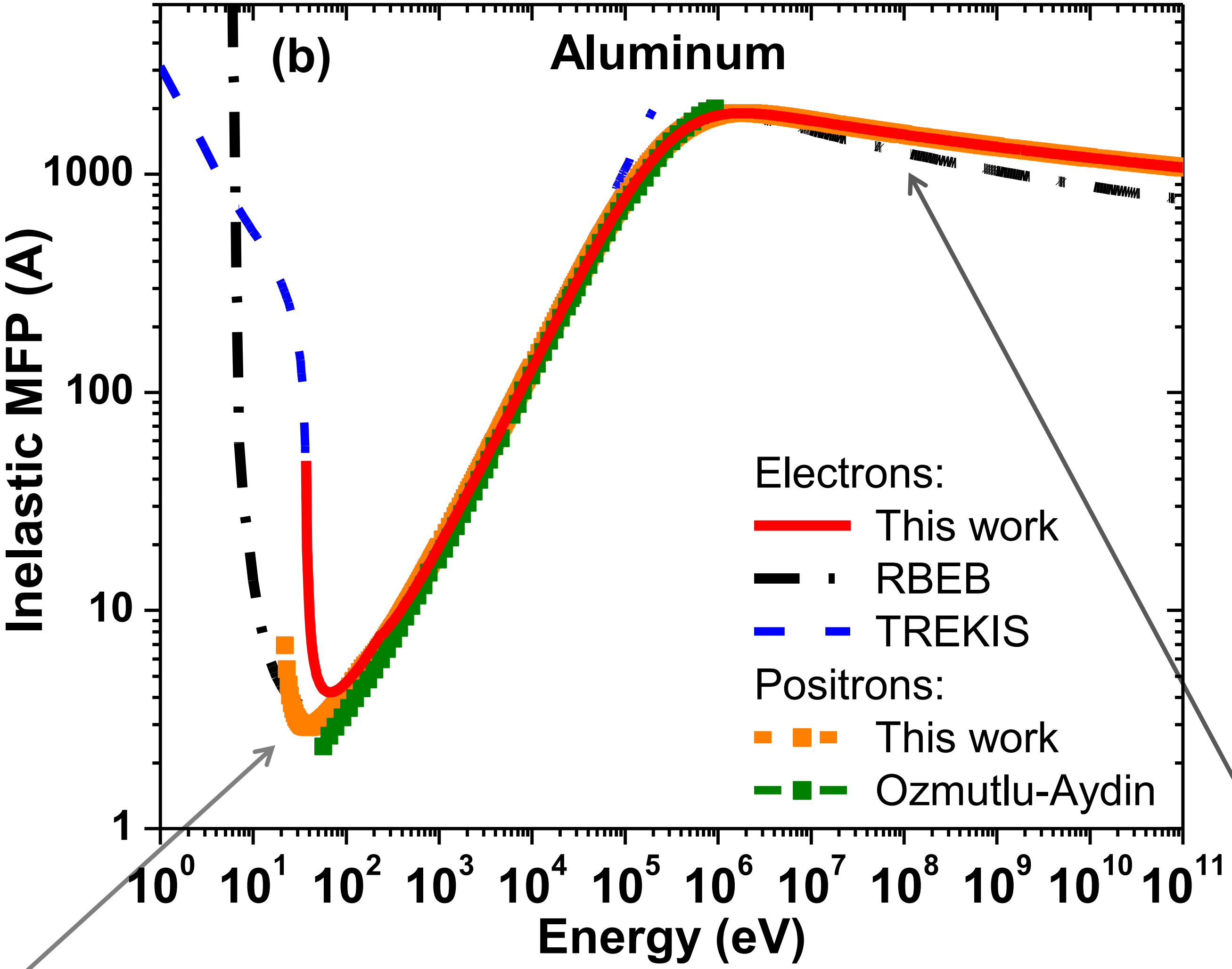
Electrons



Inelastic : impact ionization, electron-electron, electron-plasmon

Elastic : electron-atom, electron-phonon

Electrons: “inelastic” scattering

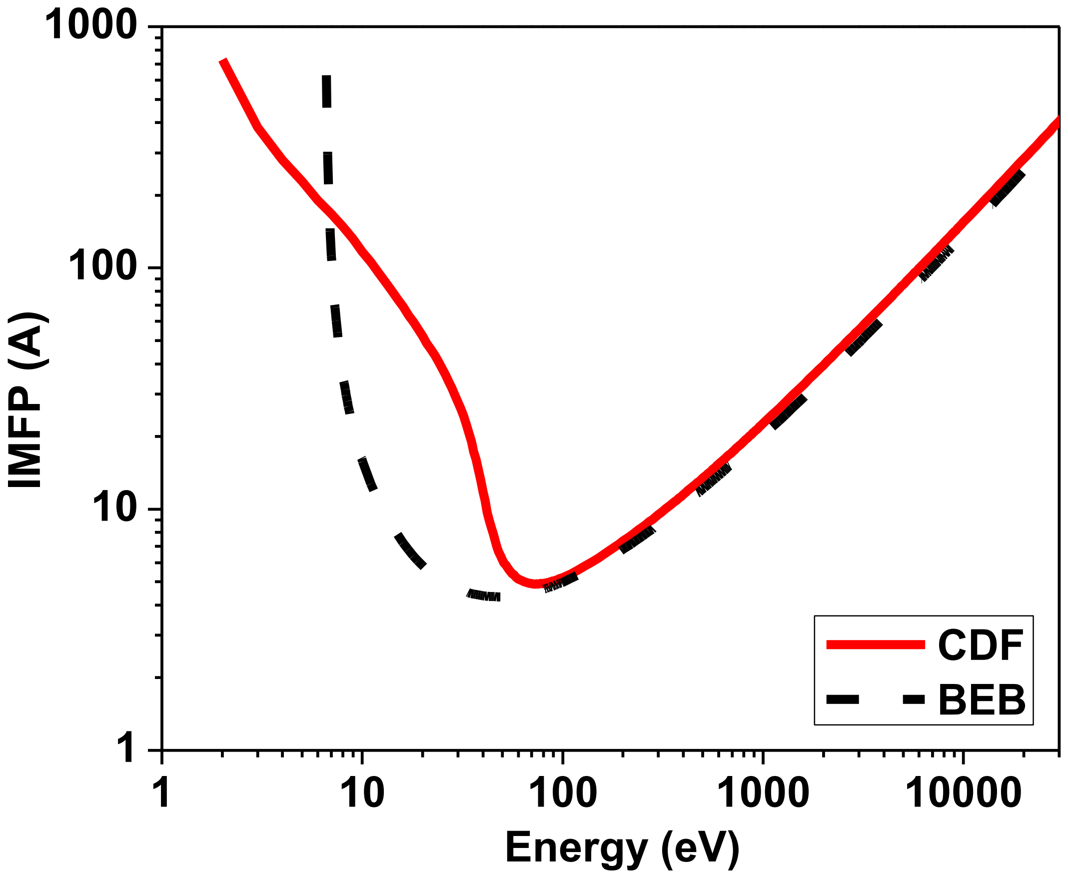


Scattering on plasmons (collective)

Scattering on individual atoms

[Medvedev, *et al*. J. Phys. D 53, 235302 (2020)]

Caution!

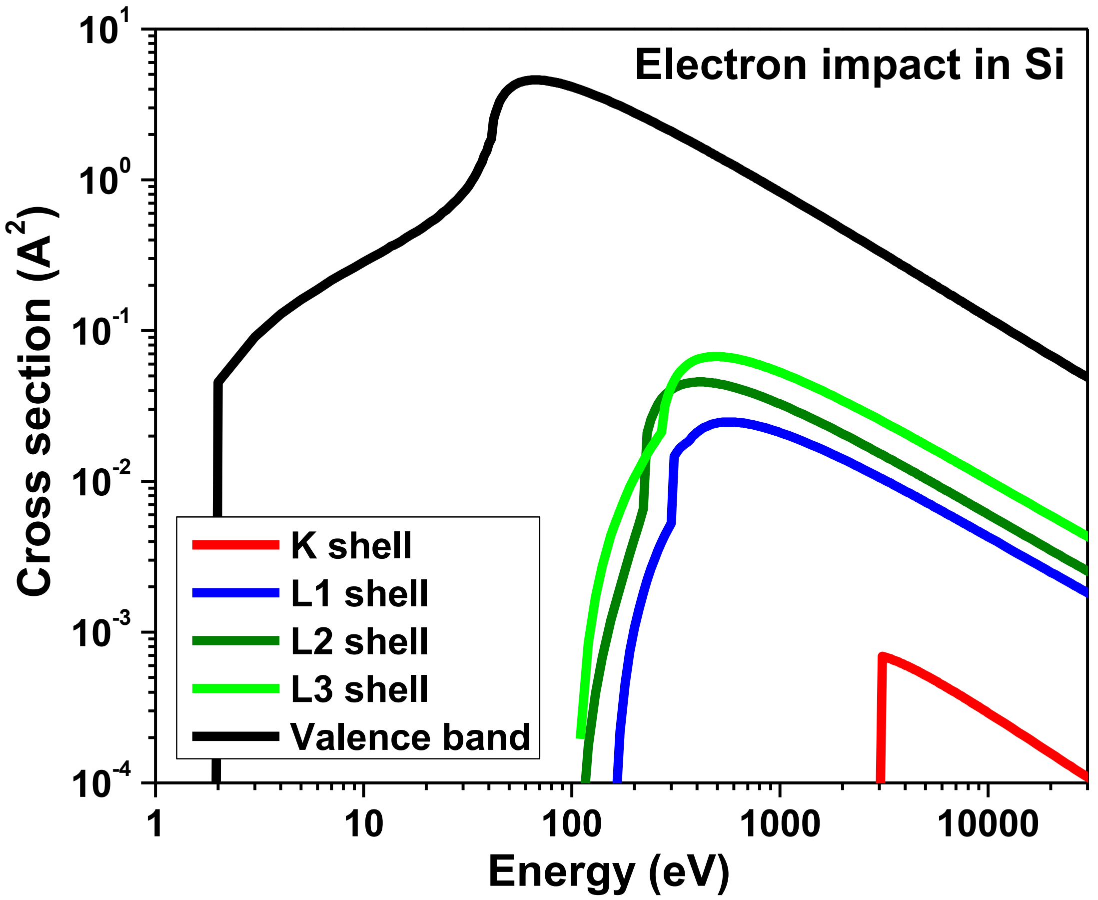


Standard MC codes often use atomic cross sections
That only works for very fast particles!

[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

Impact ionization ("inelastic")

Atomic shell choice: $P_i = \frac{\sigma_i}{\sum \sigma_i}$

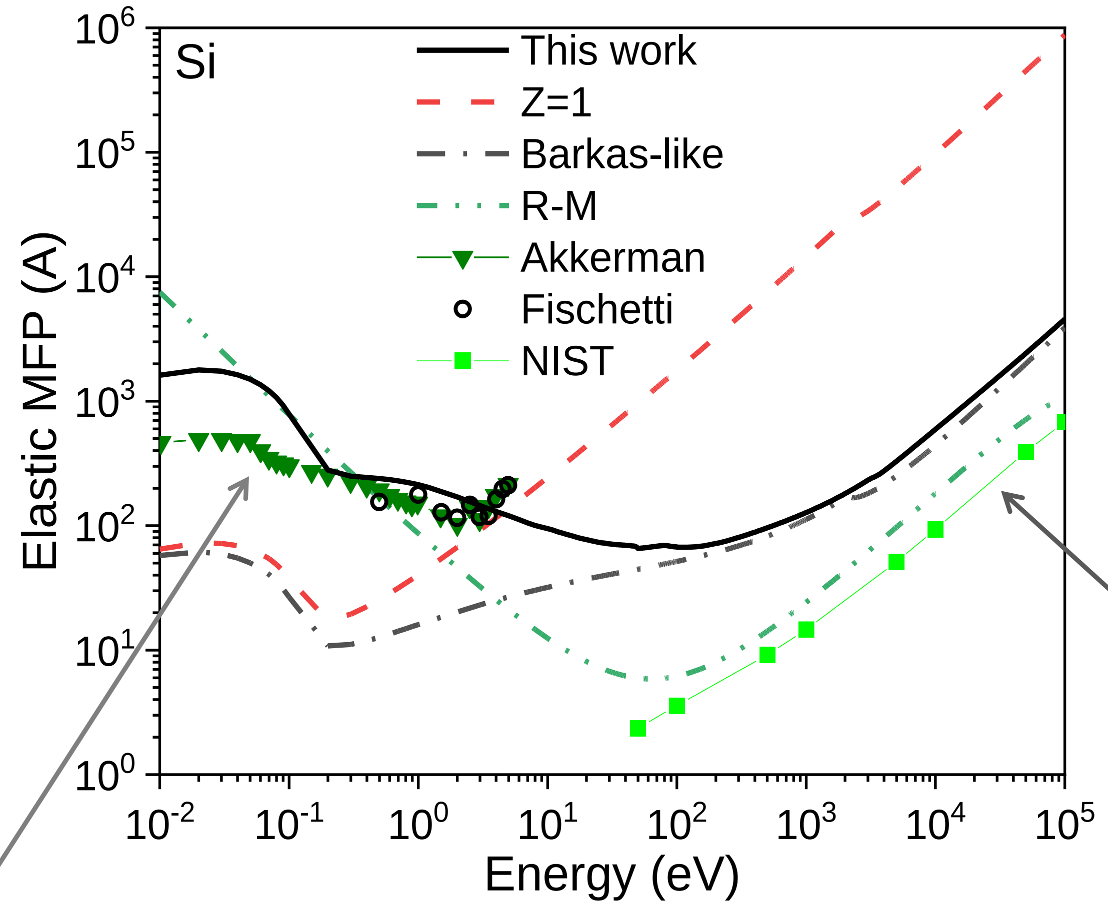


Electrons: highest shell ionization is most probable!

[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

Electrons: “elastic” scattering

$$\frac{d^2\sigma_{e-at}}{d(\hbar q)d(\hbar\omega)} = \frac{2e^2}{n_i\pi\hbar^2 v^2}\frac{1}{\hbar q}\left(1 - e^{\frac{\hbar\omega}{k_B T}}\right)^{-1}\left[Z - Z_I f_I(\tilde{q}) - N_{VB}\left(1 - \frac{1}{|\varepsilon_{VB}(\omega,\tilde{q})|}\right)\right]^2 Im\left[\frac{-1}{\tilde{\varepsilon}_{at}(\omega,q)}\right]$$



Scattering on phonons (collective)

Scattering on individual atoms

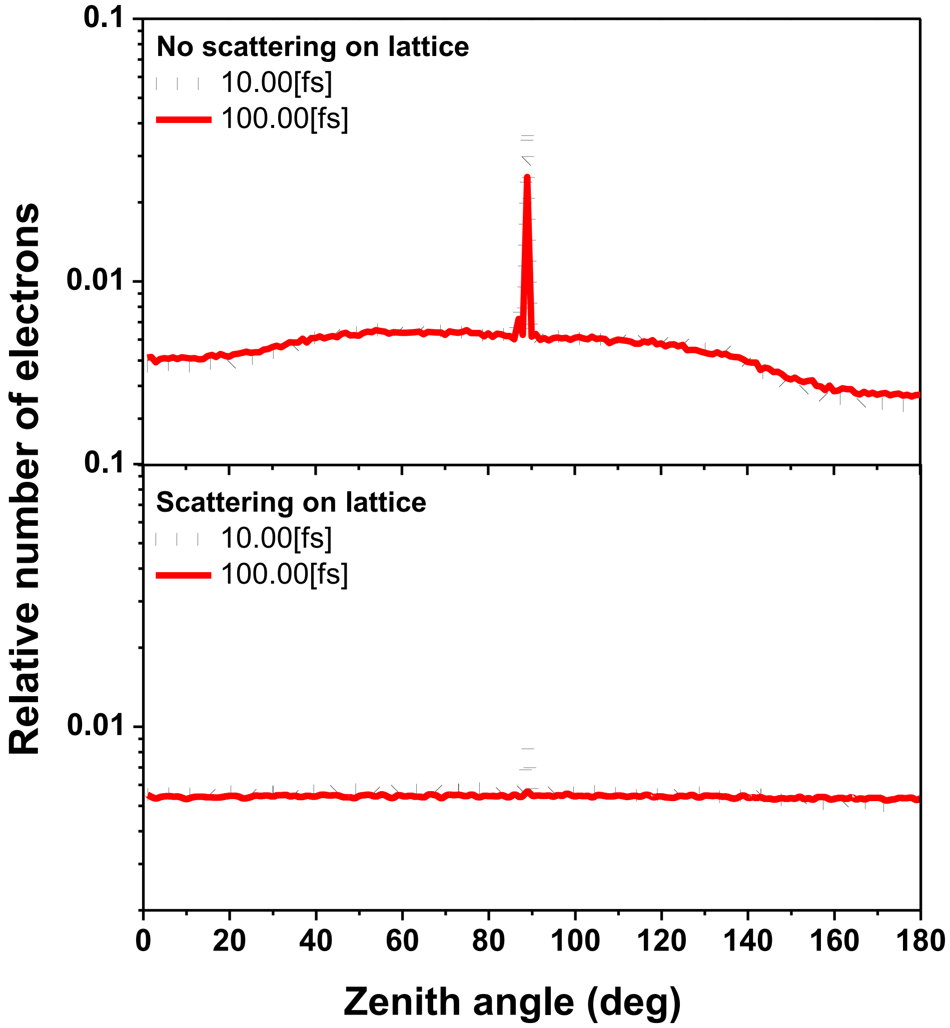
[Medvedev, *et al*. J. Appl. Phys. 137, 015903 (2025)]

Electrons: “elastic” scattering

$$E_{max} = 4E_e \frac{M_{ion} m_e}{(M_{ion} + m_e)^2} \sim 4E_e \frac{m_e}{M_{ion}}$$

Elastic scattering transfers small amount of energy,

but alters momentum a lot!



[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

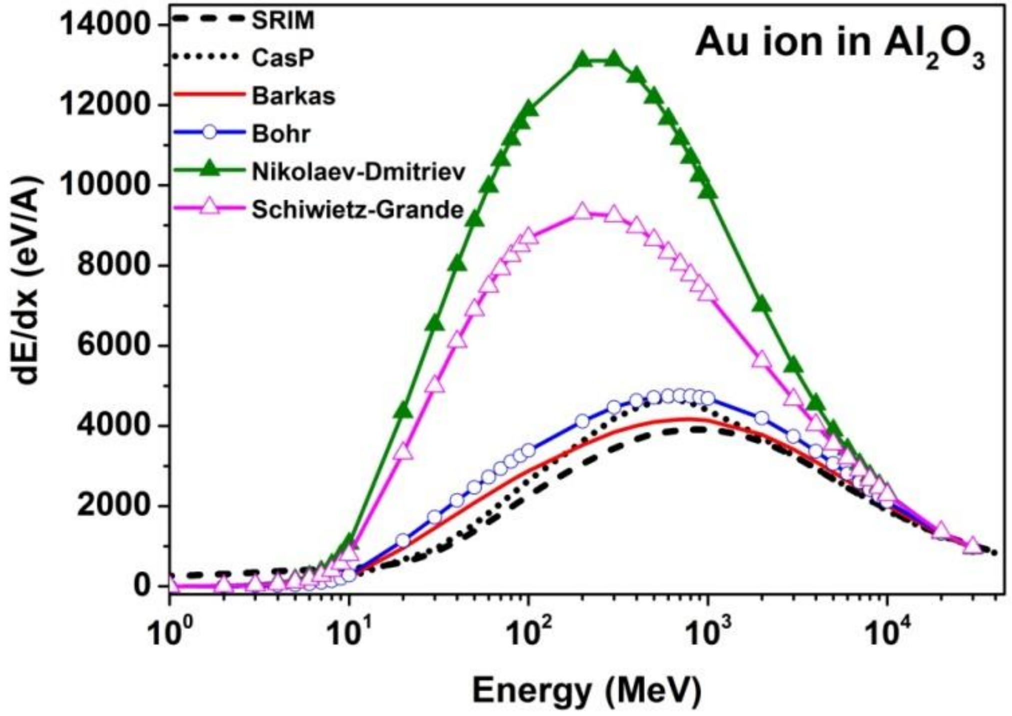
Ions

Ions are like electrons,

but heavy and highly charged

$$\frac{d^2\sigma}{d(\hbar\omega)d(\hbar q)} = \frac{2\left(Z_{eff}(v)e\right)^2}{n_{at}\pi\hbar^2 v^2}\frac{1}{\hbar q}\left[1-\exp\left(-\frac{\hbar\omega}{k_B T}\right)\right]^{-1} Im\left(\frac{-1}{\varepsilon(\omega,q)}\right)$$

Barkas formula: $Z_{eff}(v) = Z_{ion}\left[1-\exp\left(-\frac{v}{v_0}Z_{ion}^{-2/3}\right)\right]$



For ions, the Born approximation requires effective charge!

[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

Holes

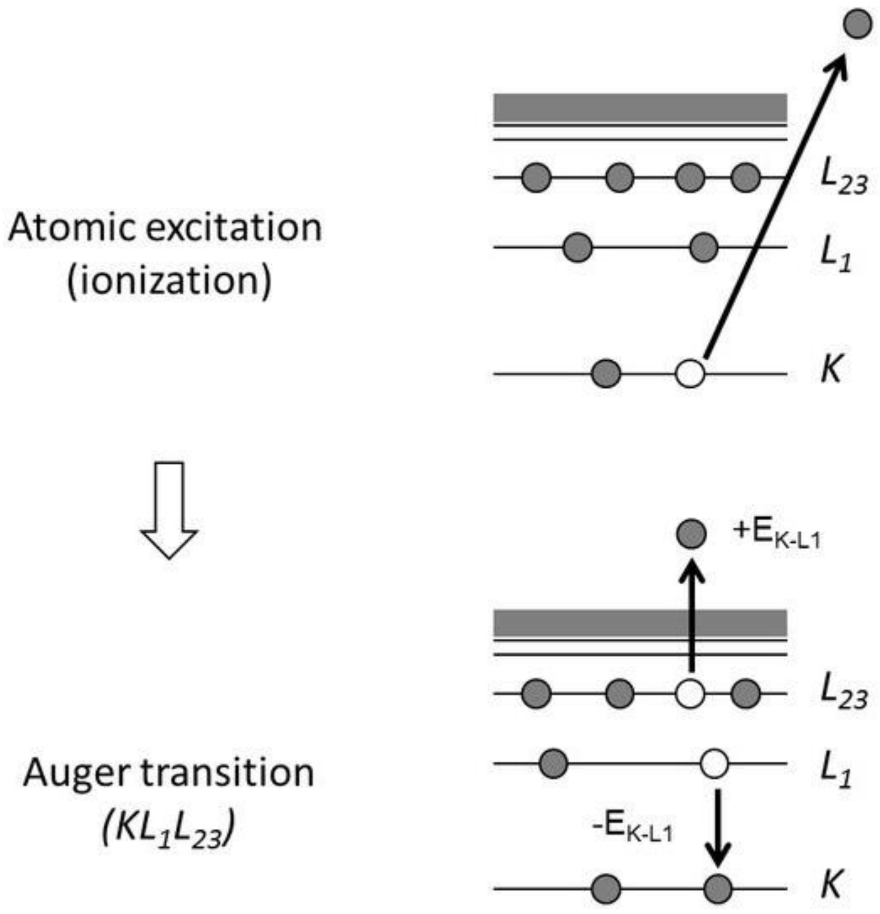
Core holes

Core holes (deep-shell):

Auger and radiative decay times

EPICS2023 database:

https://www-nds.iaea.org/epics/



Pic from: Yu-Pu Lin “Functionalization of two-dimensional nanomaterials based on graphene”

[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

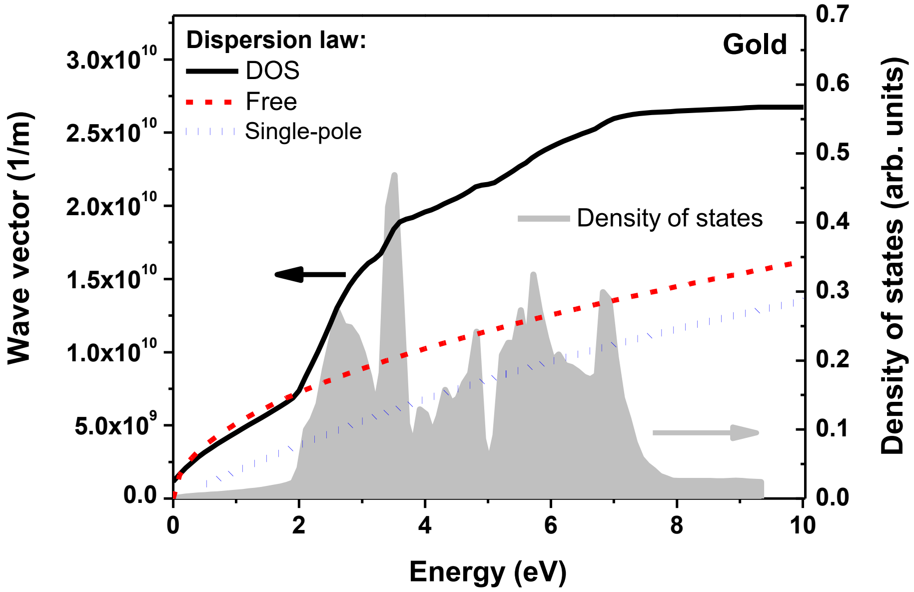
Valence holes

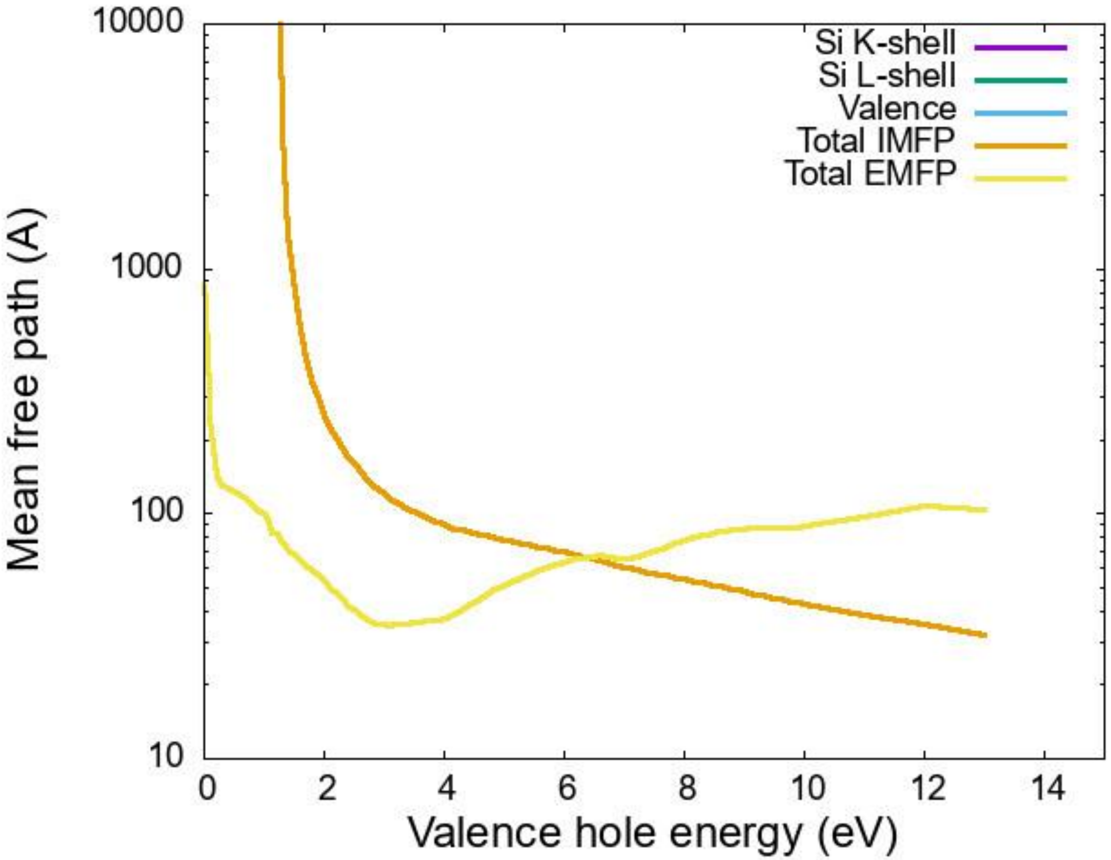
Valence holes in solids are mobile: analogous to electrons (almost)

Standard values are only valid at the top of valence band

Suggested extension – mass from DOS

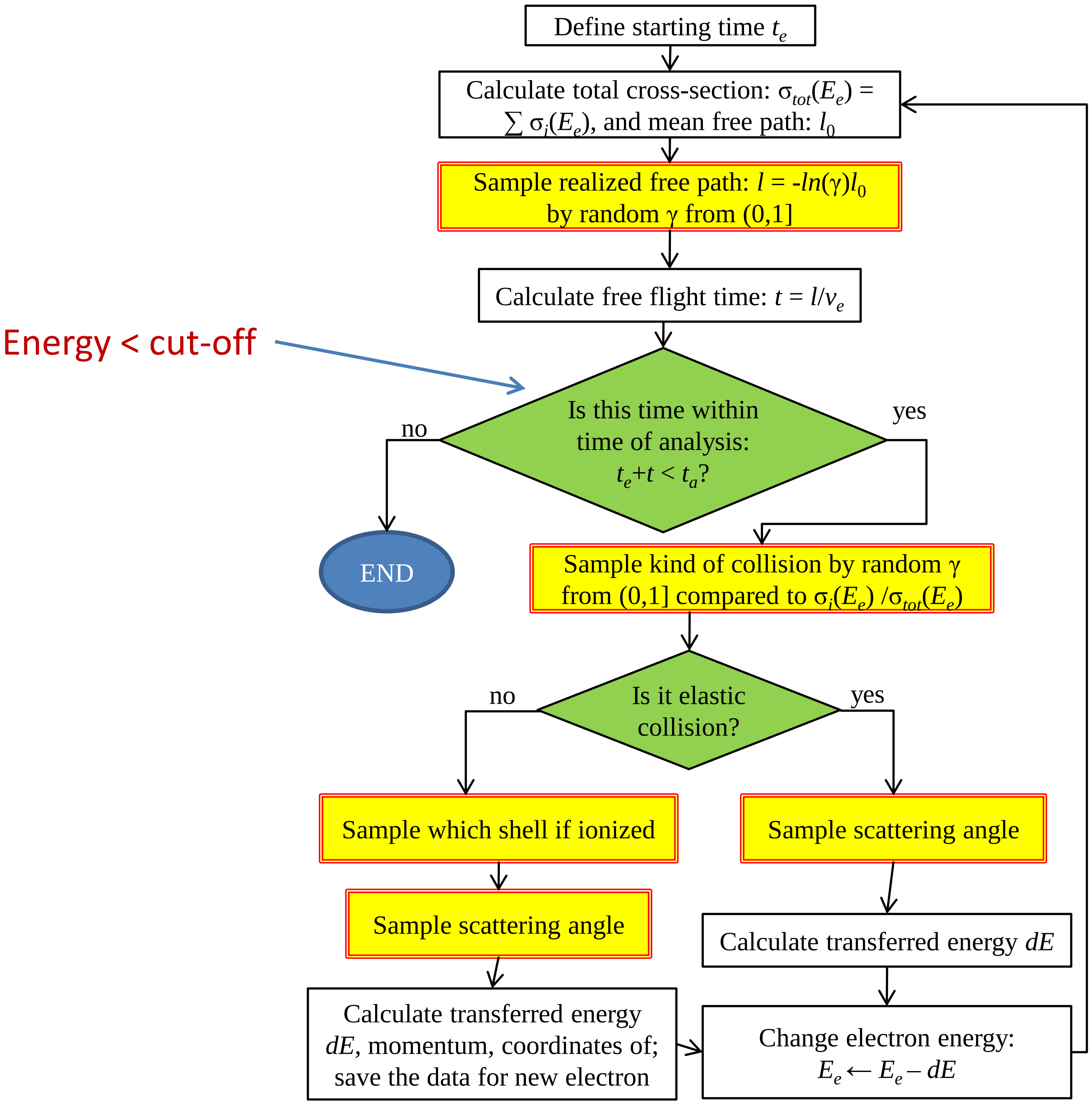
$$D(E) = \frac{s}{2\pi^2} q^2(E) \frac{dq}{dE} \qquad q(E) = \sqrt[3]{\frac{6\pi^2}{s} \int_0^E d\varepsilon \cdot D(\varepsilon)}$$





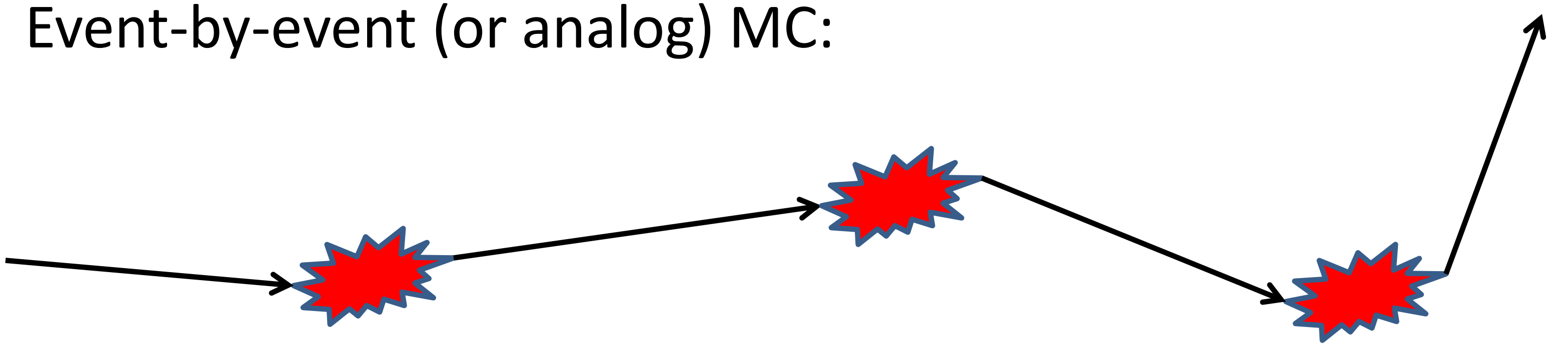
[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

MC codes and algorithms

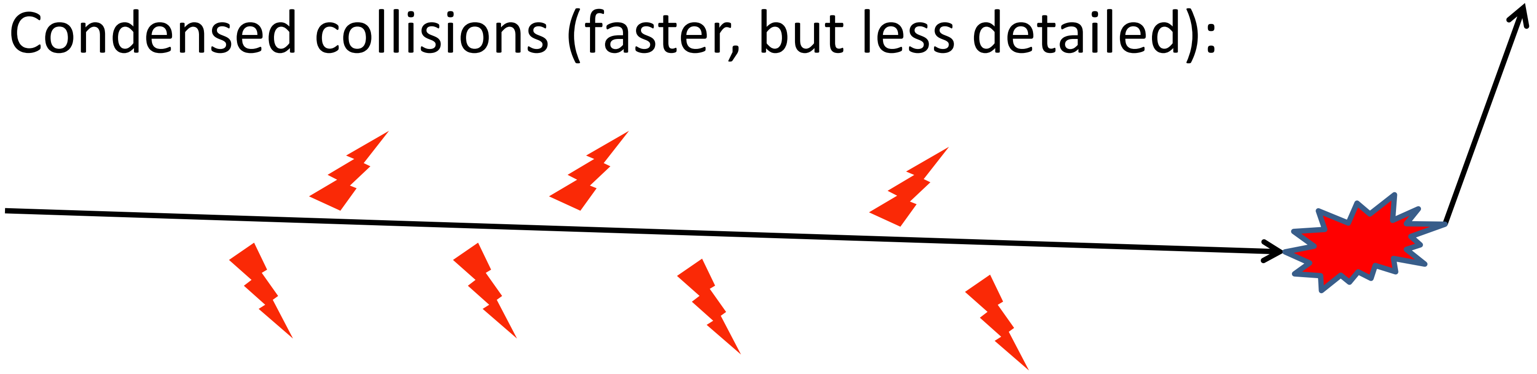

Define starting time t_e
Calculate total cross-section: $\sigma_{tot}(E_e) = \sum \sigma_i(E_e)$, and mean free path: l_0
Sample realized free path: $l = -ln(\gamma)l_0$ by random γ from (0,1]
Calculate free flight time: $t = l/v_e$
Energy < cut-off
Is this time within time of analysis: $t_e+t < t_a$?
no
yes
END
Sample kind of collision by random γ from (0,1] compared to $\sigma_i(E_e) / \sigma_{tot}(E_e)$
Is it elastic collision?
no
yes
Sample which shell if ionized
Sample scattering angle
Sample scattering angle
Calculate transferred energy dE
Calculate transferred energy dE, momentum, coordinates of; save the data for new electron
Change electron energy: $E_e \leftarrow E_e - dE$

MC algorithms

Event-by-event (or analog) MC:



Condensed collisions (faster, but less detailed):



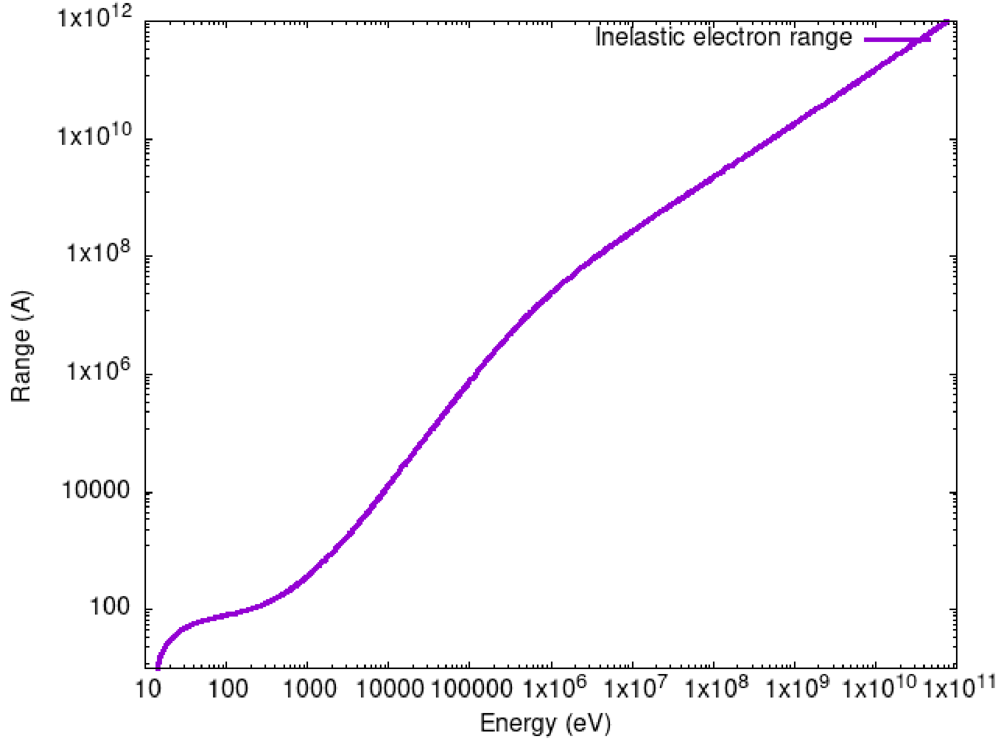
For combined simulations, only event-by-event works

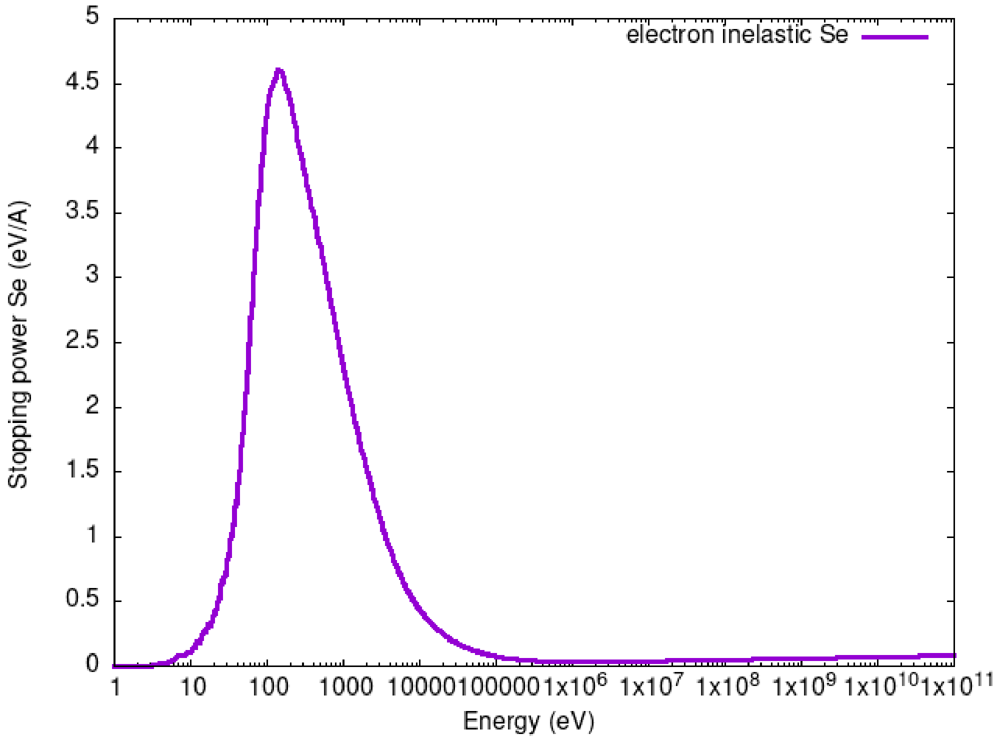
Typical MC outputs: range

Range: total distance until coming to a “stop” (down to 10 eV):

$$R_0(E) = \int_{10\,\mathrm{eV}}^{E} \frac{dE'}{S(E')}$$

$$S_e = -\frac{dE}{dx} = n_{at} \int_{E_{min}}^{E_{max}} \int_{q_-}^{q_+} \hbar\omega \frac{d^2\sigma}{d(\hbar\omega)d(\hbar q)} d(\hbar\omega)d(\hbar q)$$





Typical MC outputs: cumulative dose

Energy cut off is often used (beware, may be unphysical!)

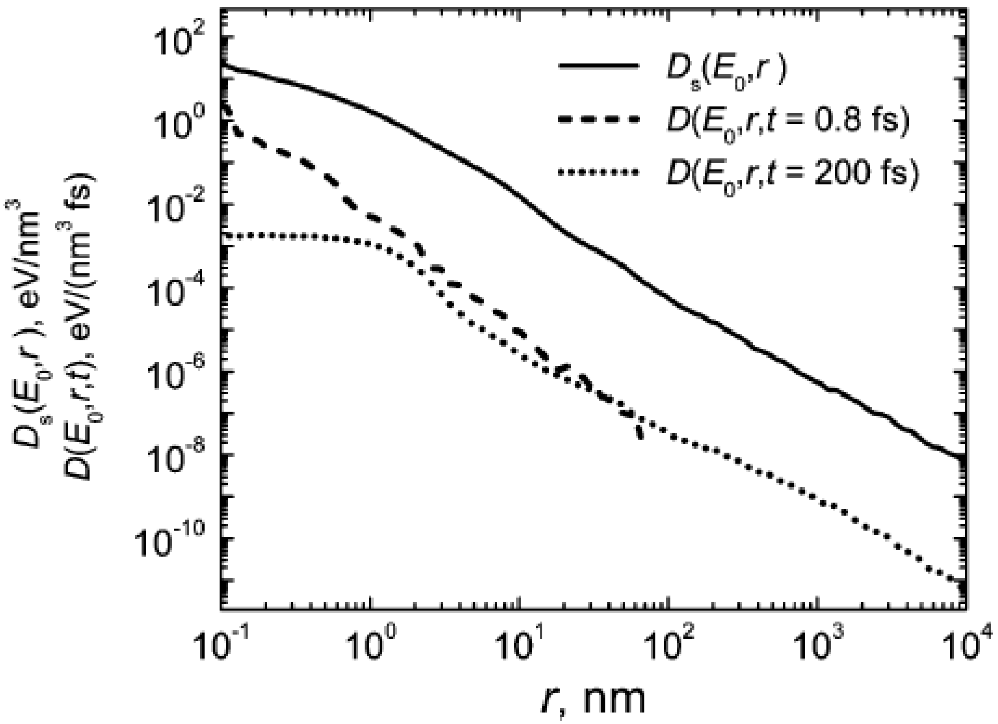


Fig. 2. Differential spatial deposited energy distributions at two discrete times for 100 MeV/amu Ne ions. The upper line is the energy deposition distribution integrated over time.

Cumulative dose does not correspond to real dose at any time instant

Murat, Akkerman, Barak, IEEE Transactions on Nuclear Science 55, 2113 (2008)

MC codes

Standard MC codes for radiation transport:

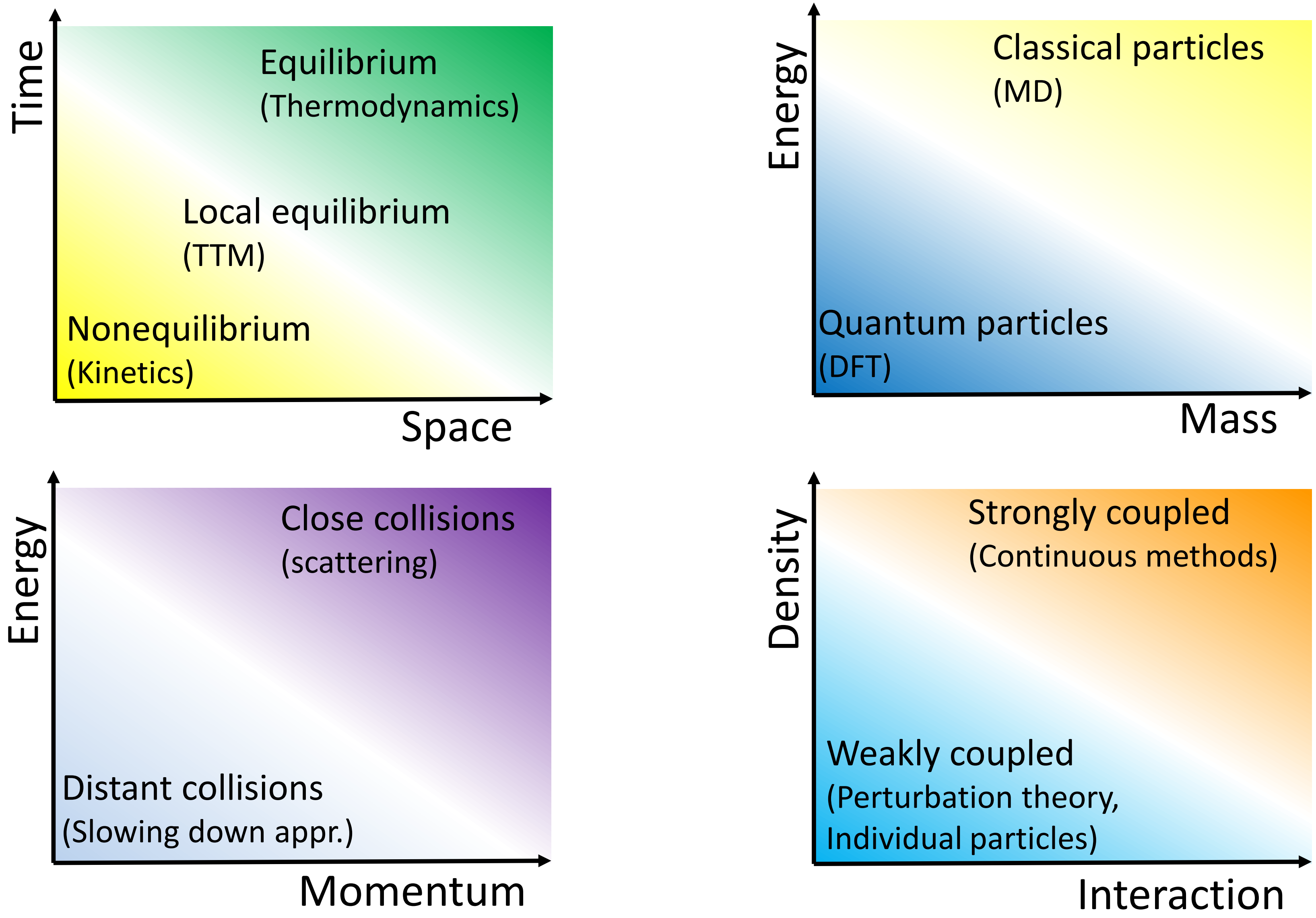
GEANT4:	https://geant4.web.cern.ch/
FLUKA:	http://www.fluka.org/fluka.php
PENELOPE:	http://pypenelope.sourceforge.net
PHITS:	https://phits.jaea.go.jp/
MCNP:	https://mcnp.lanl.gov/
TART:	http://redcullen1.net/homepage.new/tart2016.htm

Our MC codes:

TREKIS-3:	https://github.com/N-Medvedev/TREKIS-3
TREKIS-4:	https://github.com/N-Medvedev/TREKIS-4

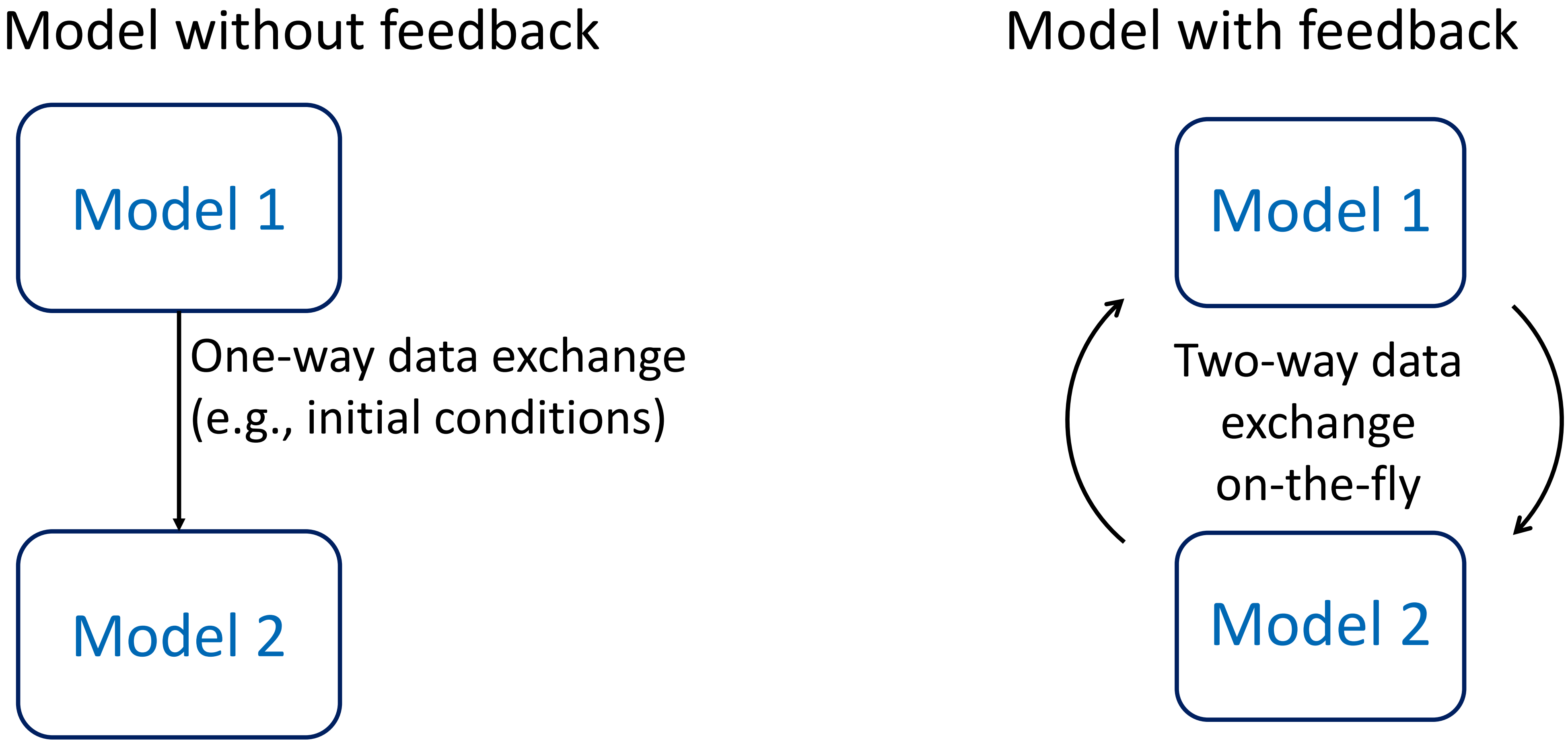
Combining MC codes with others

Combined / hybrid / multiscale approaches



Apostolova *et al.*, “*Tools for investigating electronic excitation: experiment and multi-scale modelling*”
https://doi.org/10.20868/UPM.book.69109

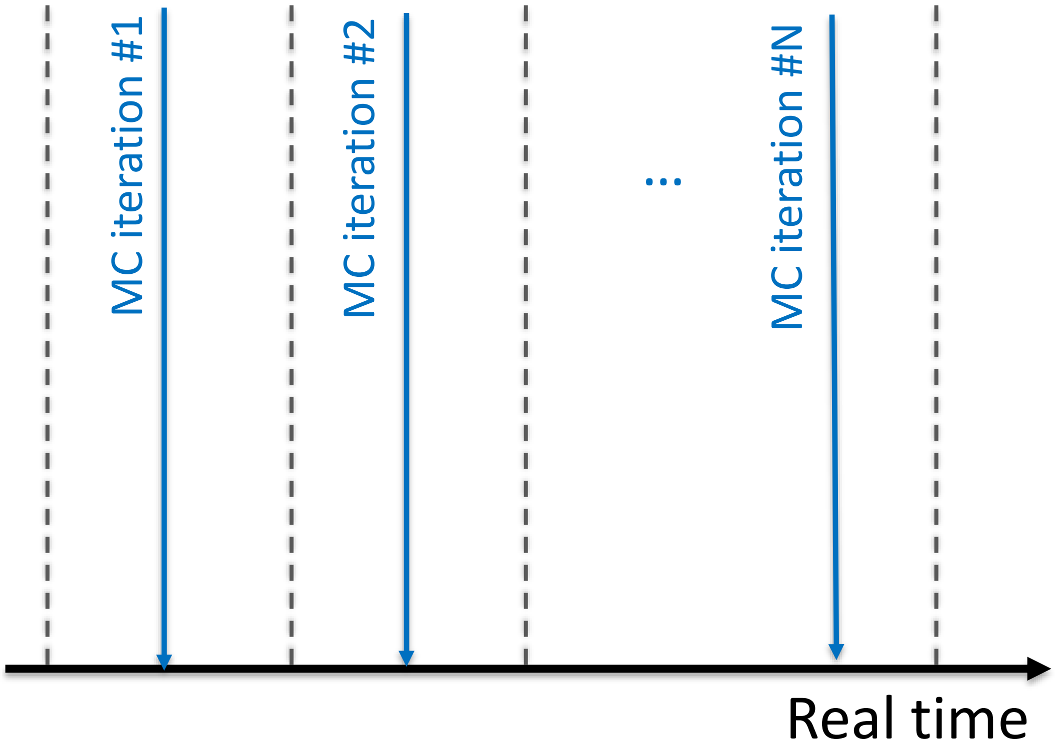
Combining MC codes



Apostolova *et al.*, *"Tools for investigating electronic excitation: experiment and multi-scale modelling"*
https://doi.org/10.20868/UPM.book.69109

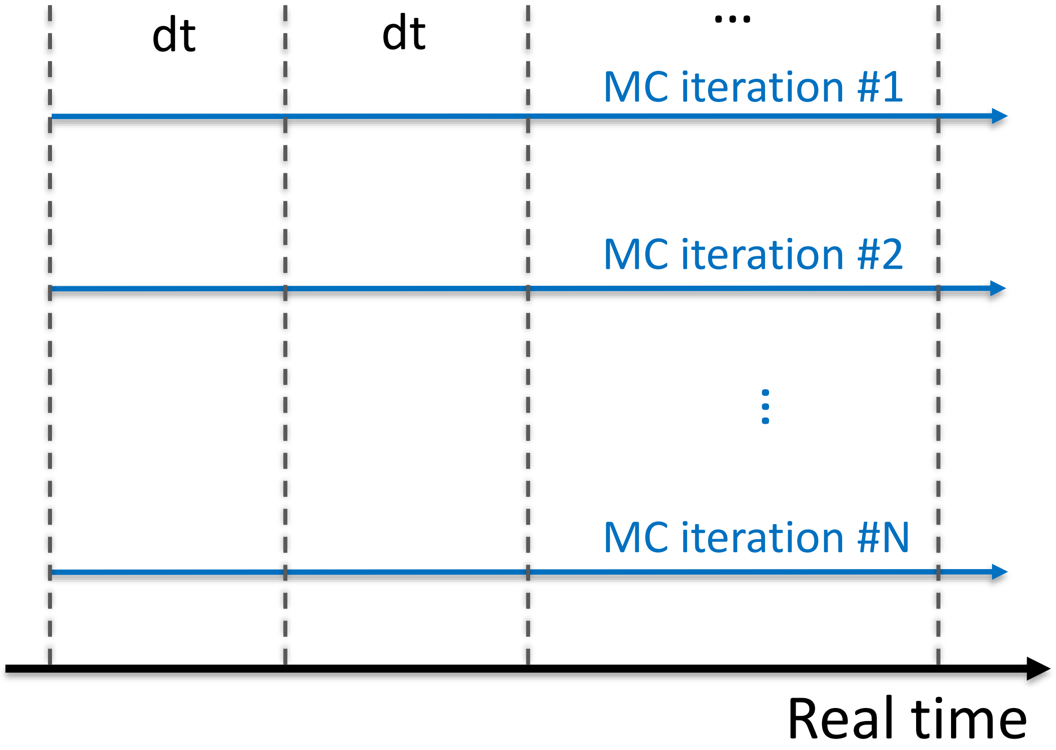
Combining MC codes

Standard MC method
(combining without feedback)



Output: distribution at final time

MC methods as part of hybrids with feedback



Output: distribution at each timestep

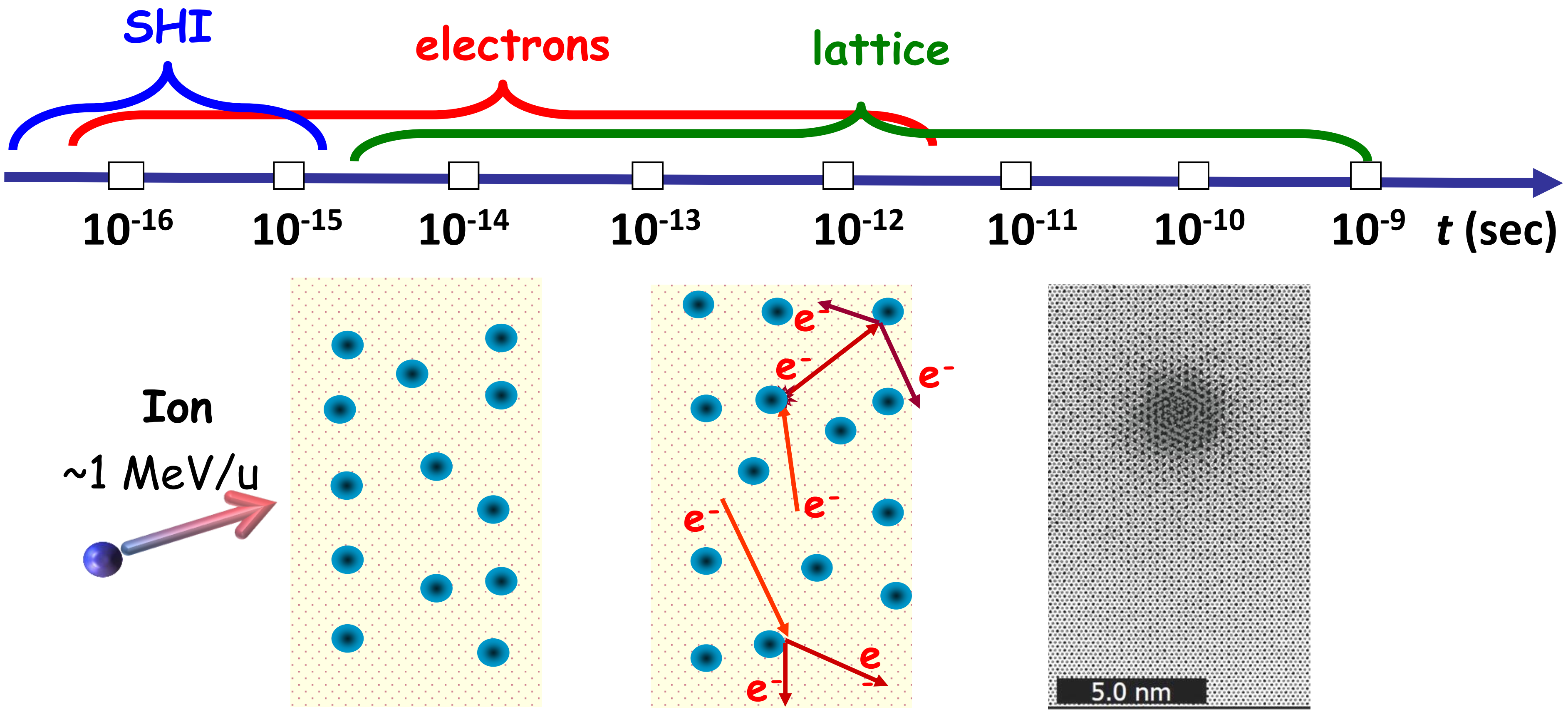
Apostolova *et al.*, *"Tools for investigating electronic excitation: experiment and multi-scale modelling"*
https://doi.org/10.20868/UPM.book.69109

Example: Models without feedback

Example: ion beam

Time-Resolved Electron Kinetics in SHI-Irradiated Solids

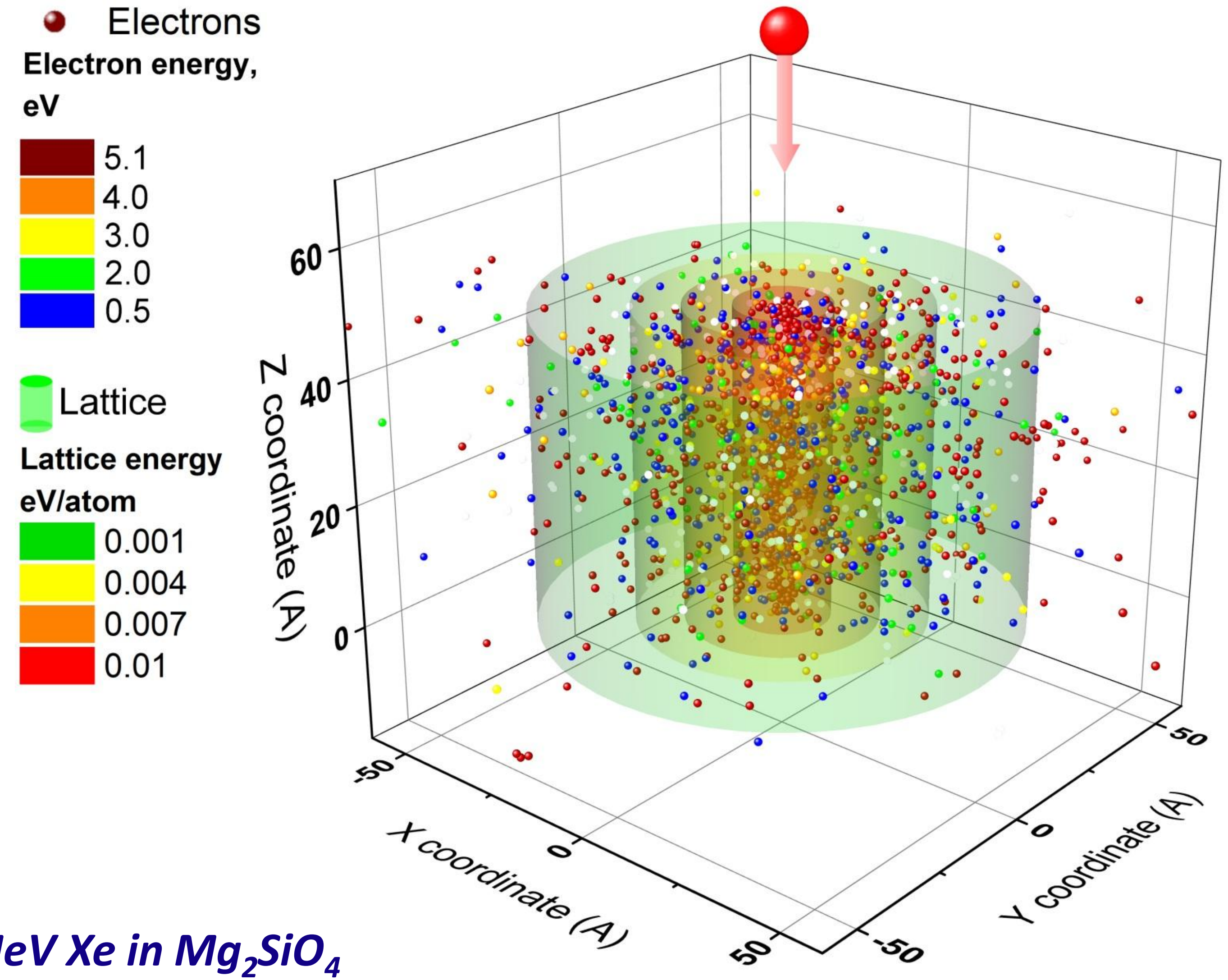
Event-by-event Monte Carlo code for simulation of



[R. Rymzhanov]

https://github.com/N-Medvedev/TREKIS-3

Example: ion beam

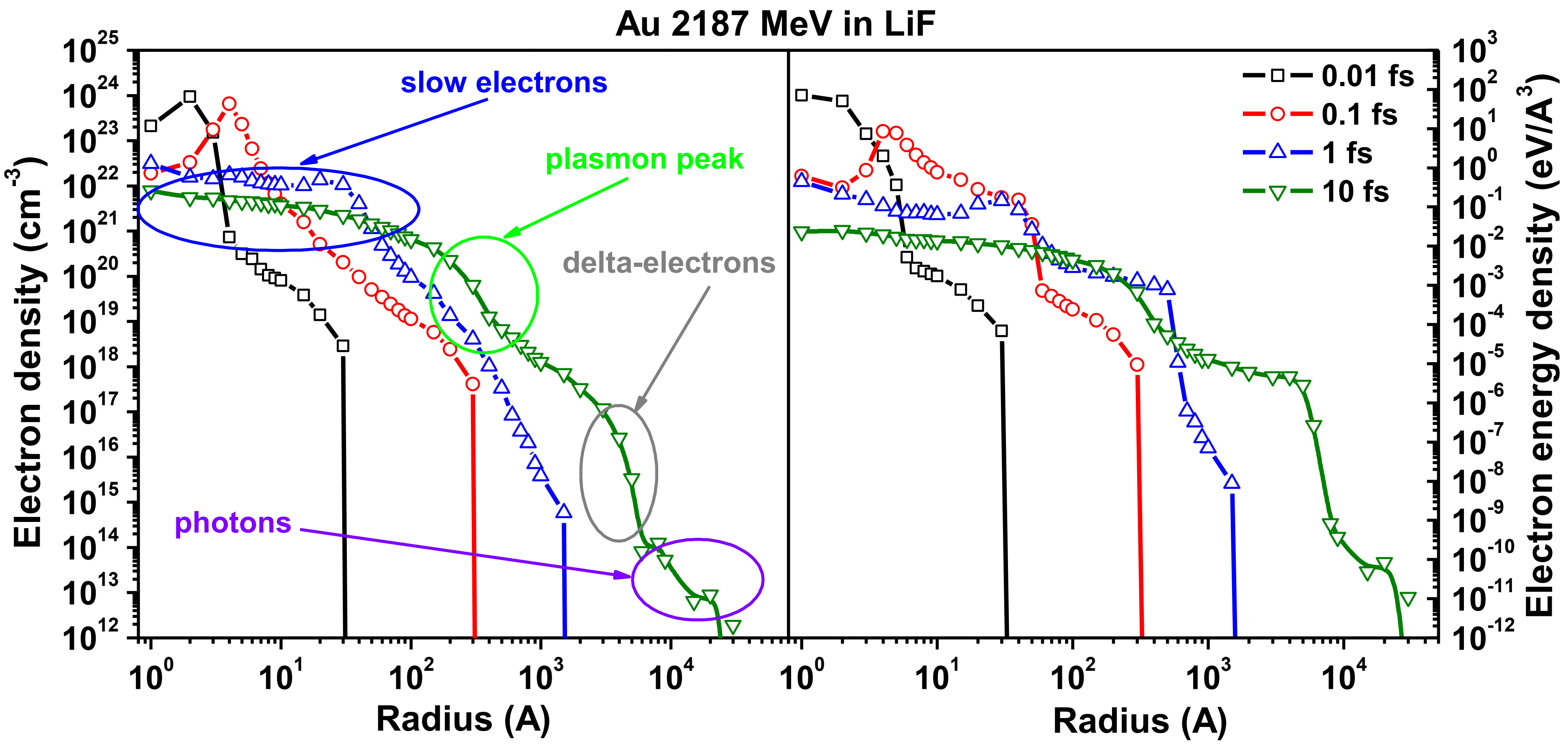


2 fs after 130 MeV Xe in Mg_2SiO_4

Pic: courtesy of R. Rymzhanov

https://github.com/N-Medvedev/TREKIS-3

Example: ion beam



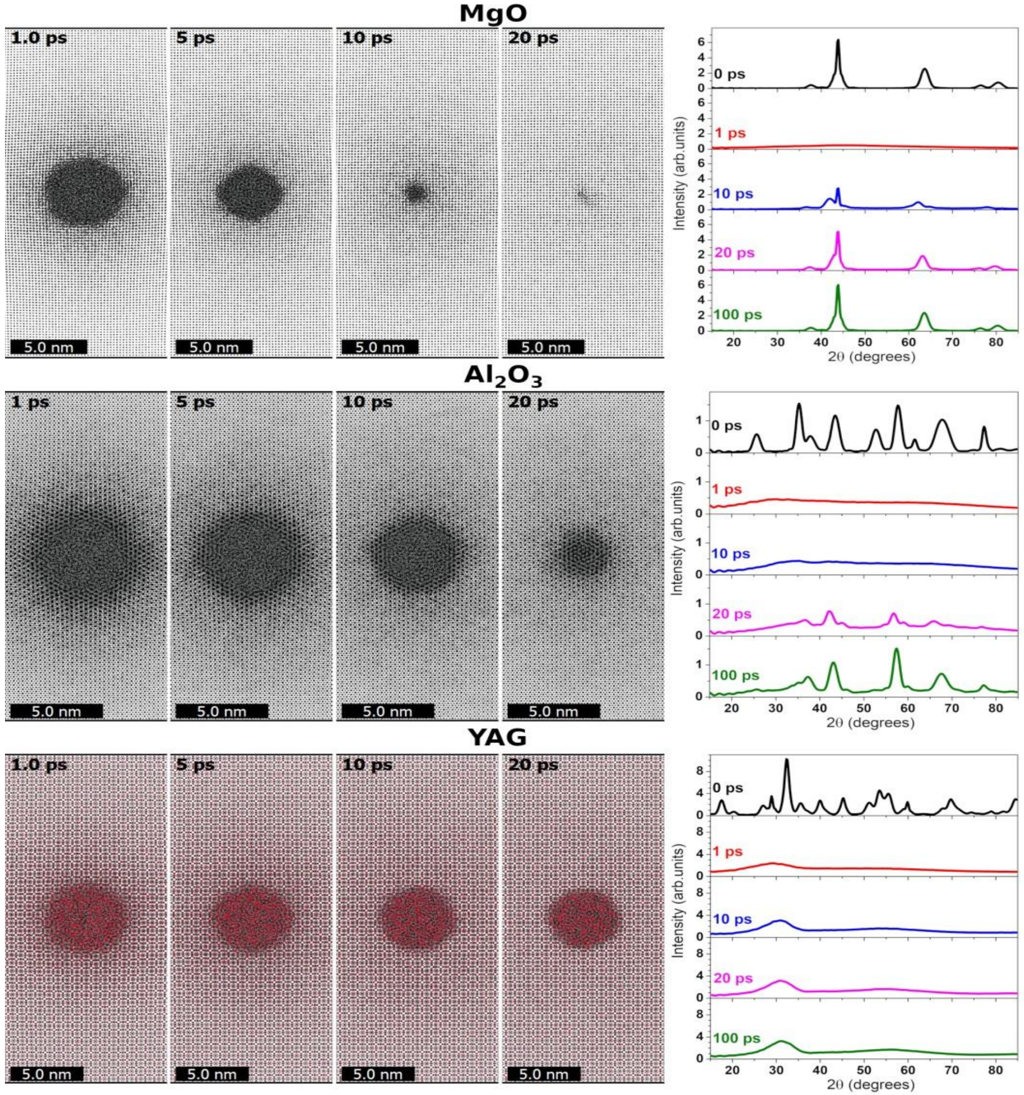
Front waves: ballistic transport

Slow electrons near track center: diffusive behavior

[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

Example: ion beam

TREKIS[3] **output ⇒ input for** LAMMPS



MgO — **Complete recrystallization**

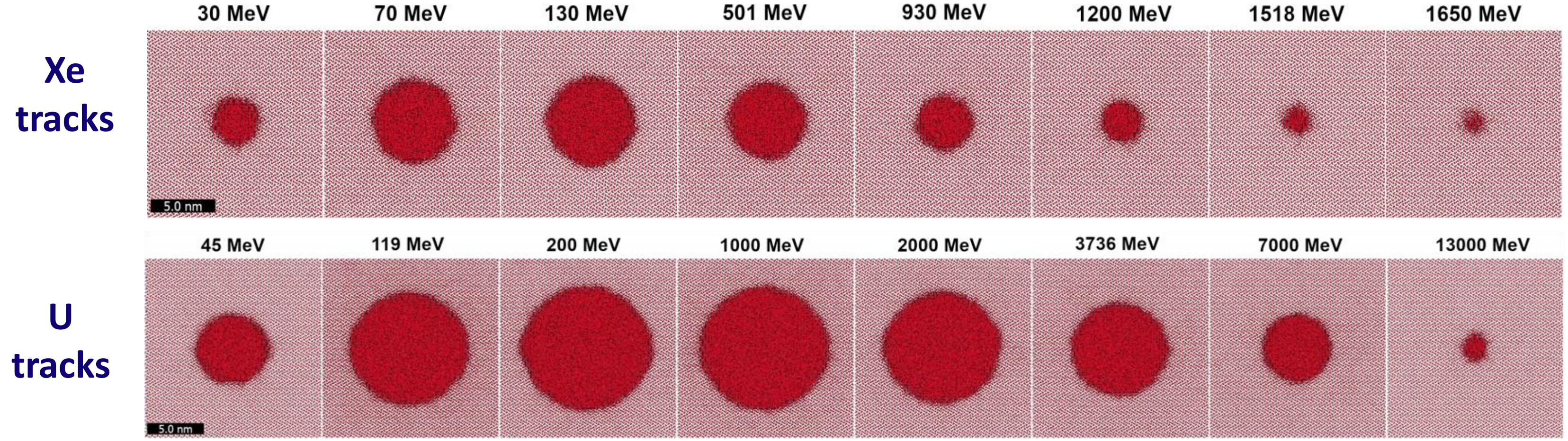
Al_2O_3 — **Partial recrystallization**

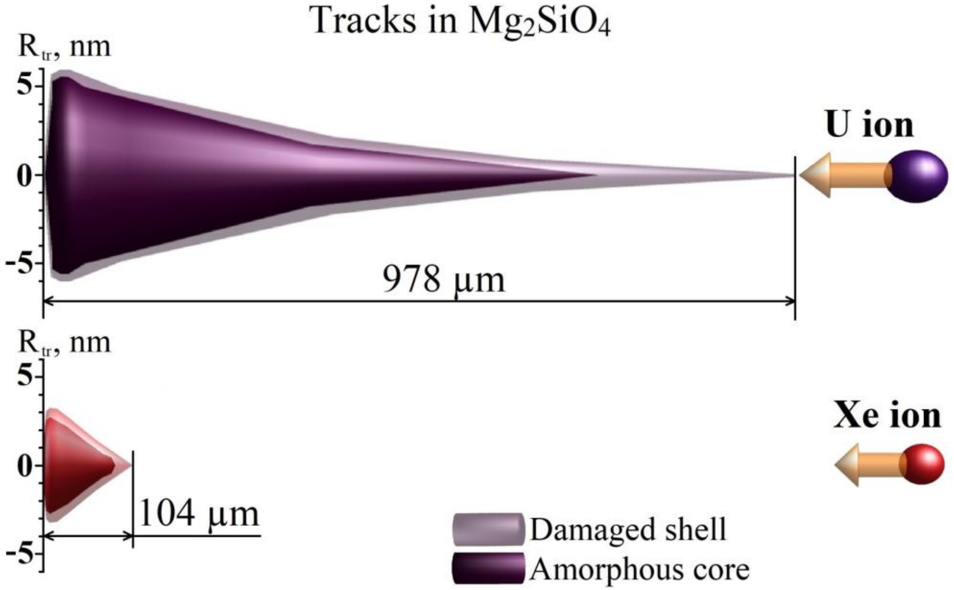
YAG — **No recrystallization**

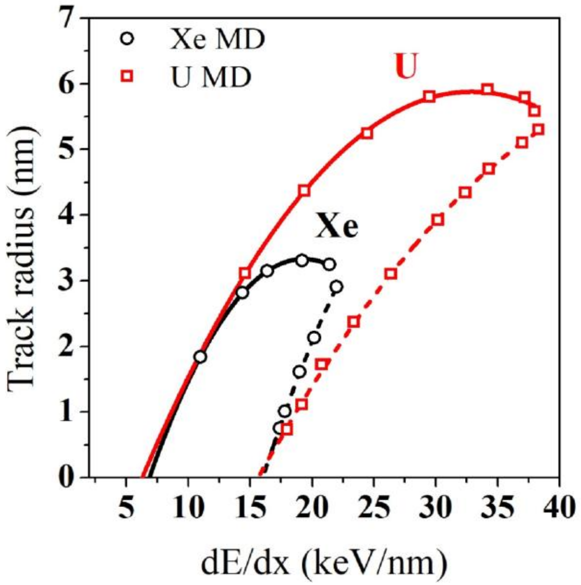
[Rymzhanov *et al.*, Scientific Reports 9, 3837 (2019)]

Example: ion beam

TREKIS[3] **output ⇒ input for** LAMMPS



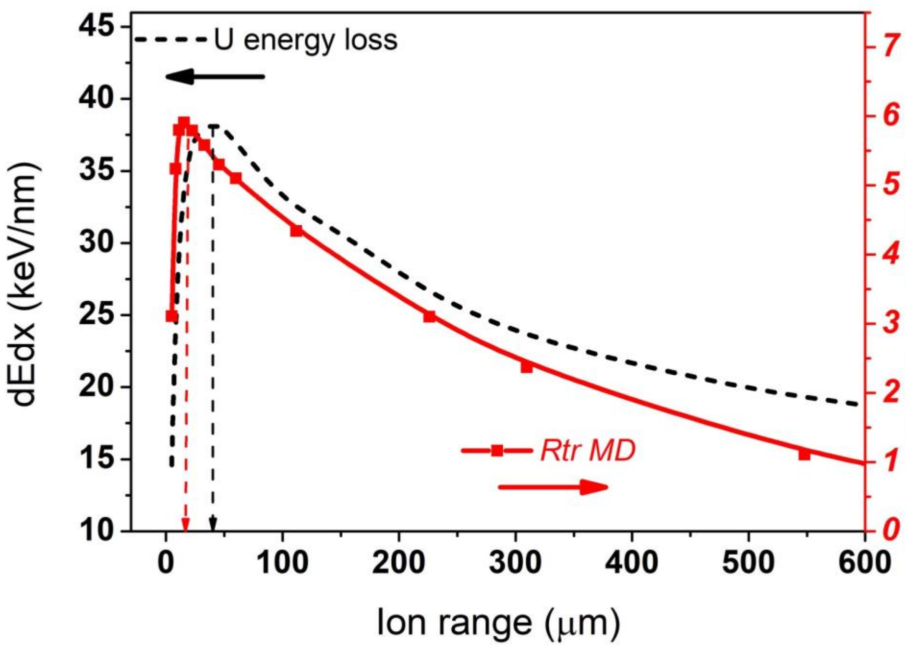


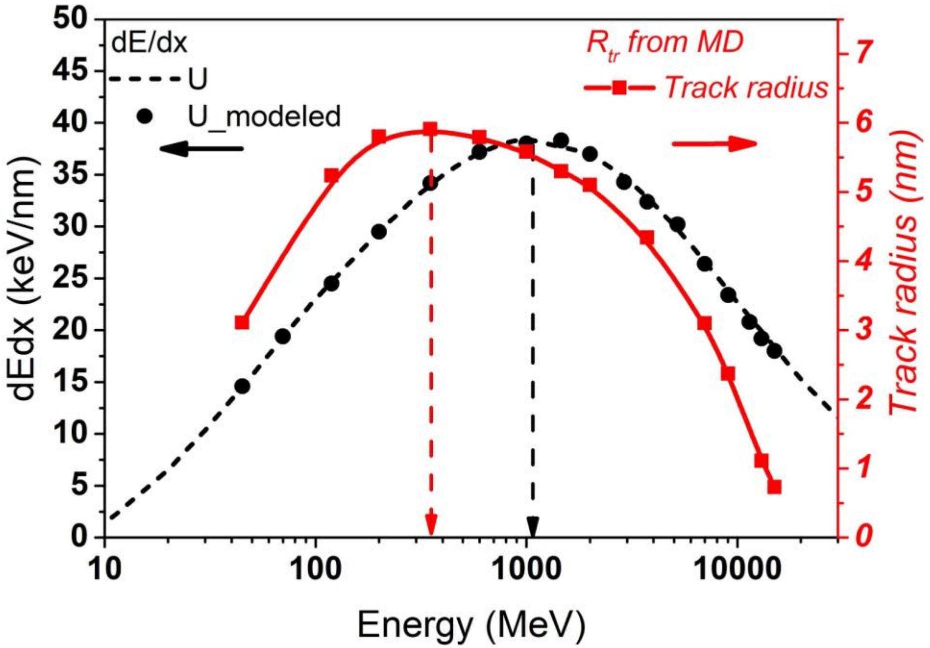


[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

Example: ion beam

TREKIS³ output ⇒ input for LAMMPS





Maximal damage does not coincide with the maximal dose

Knowing the dose from MC is not sufficient to predict damage!

[Medvedev *et al.*, J. Appl. Phys. 133, 100701 (2023)]

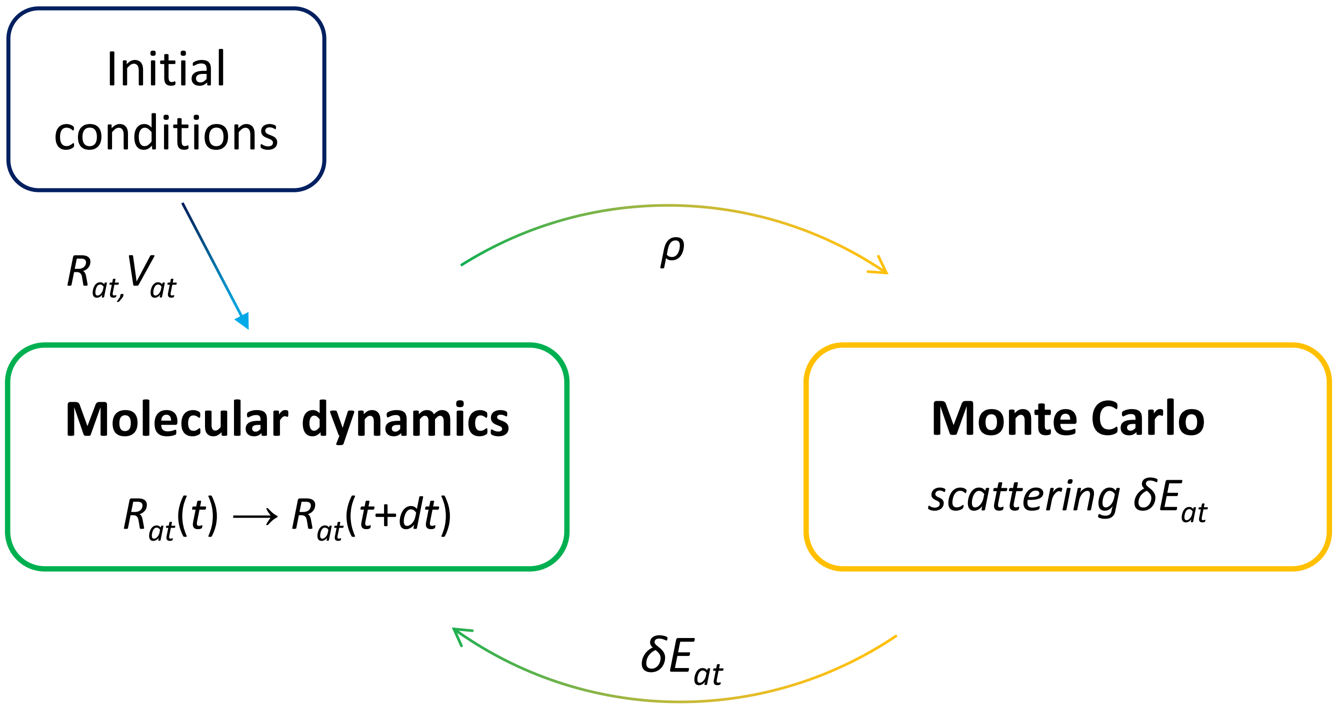
Example: Models with feedback

Example: ion beam

TREKIS⁴

Time-REsolved Kinetics in Irradiated Solids

Generalization of TTM-MD to nonequilibrium electrons



Electrons:

Transport MC

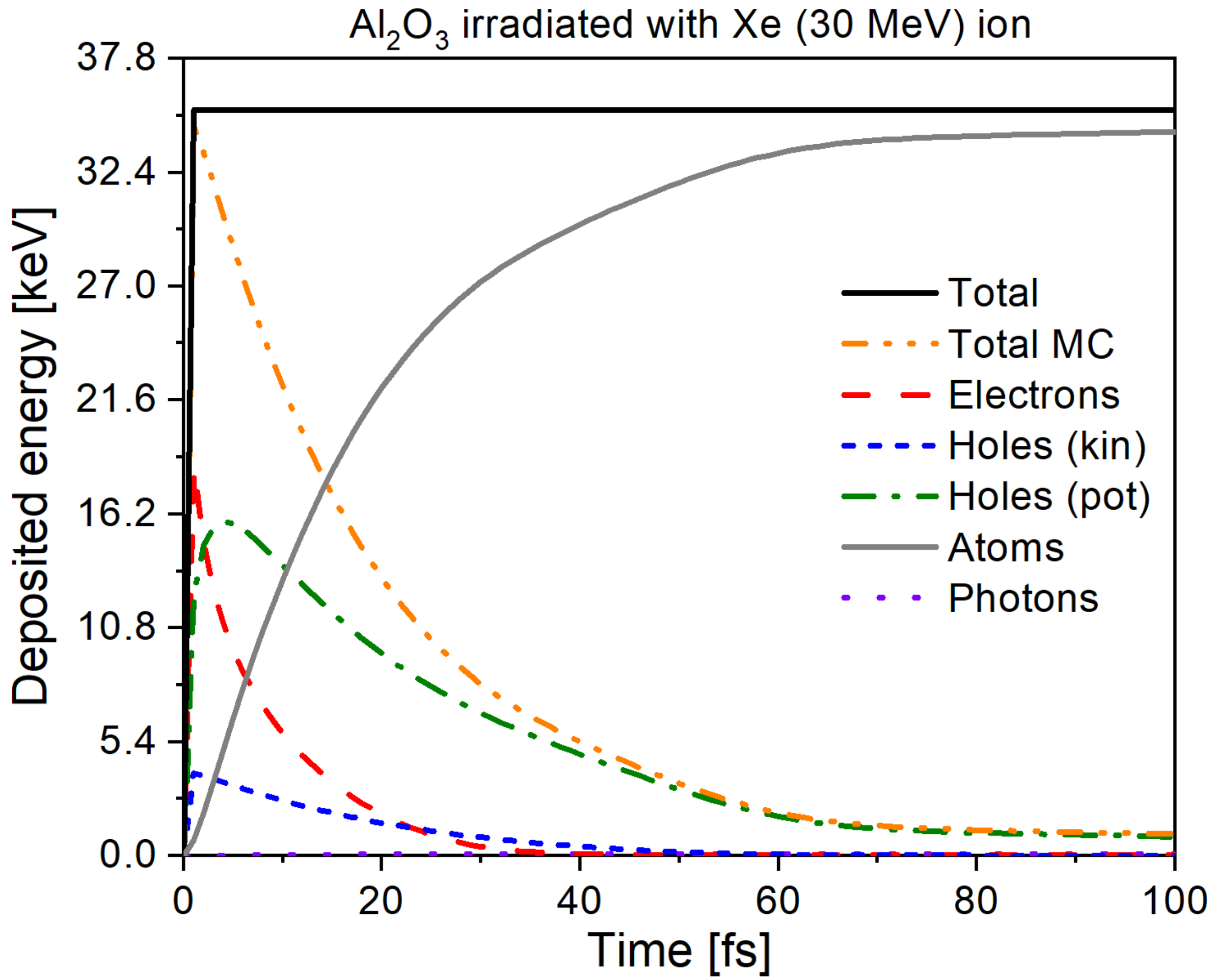
Atoms:

Classical MD

On-the-fly exchange of information between MC and MD

https://github.com/N-Medvedev/TREKIS-4

Example: ion beam

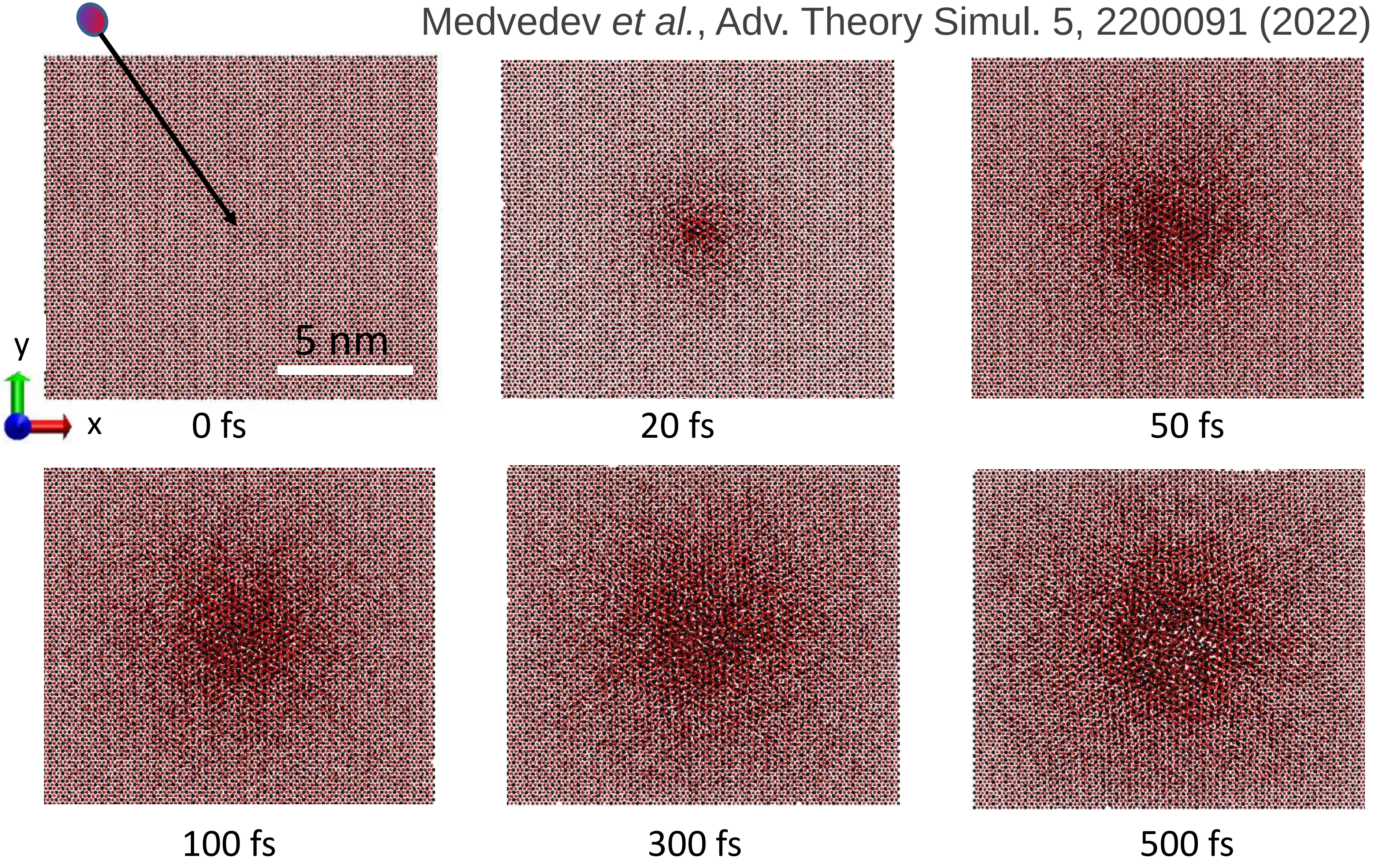


On-the-fly exchange of information between MC and MD

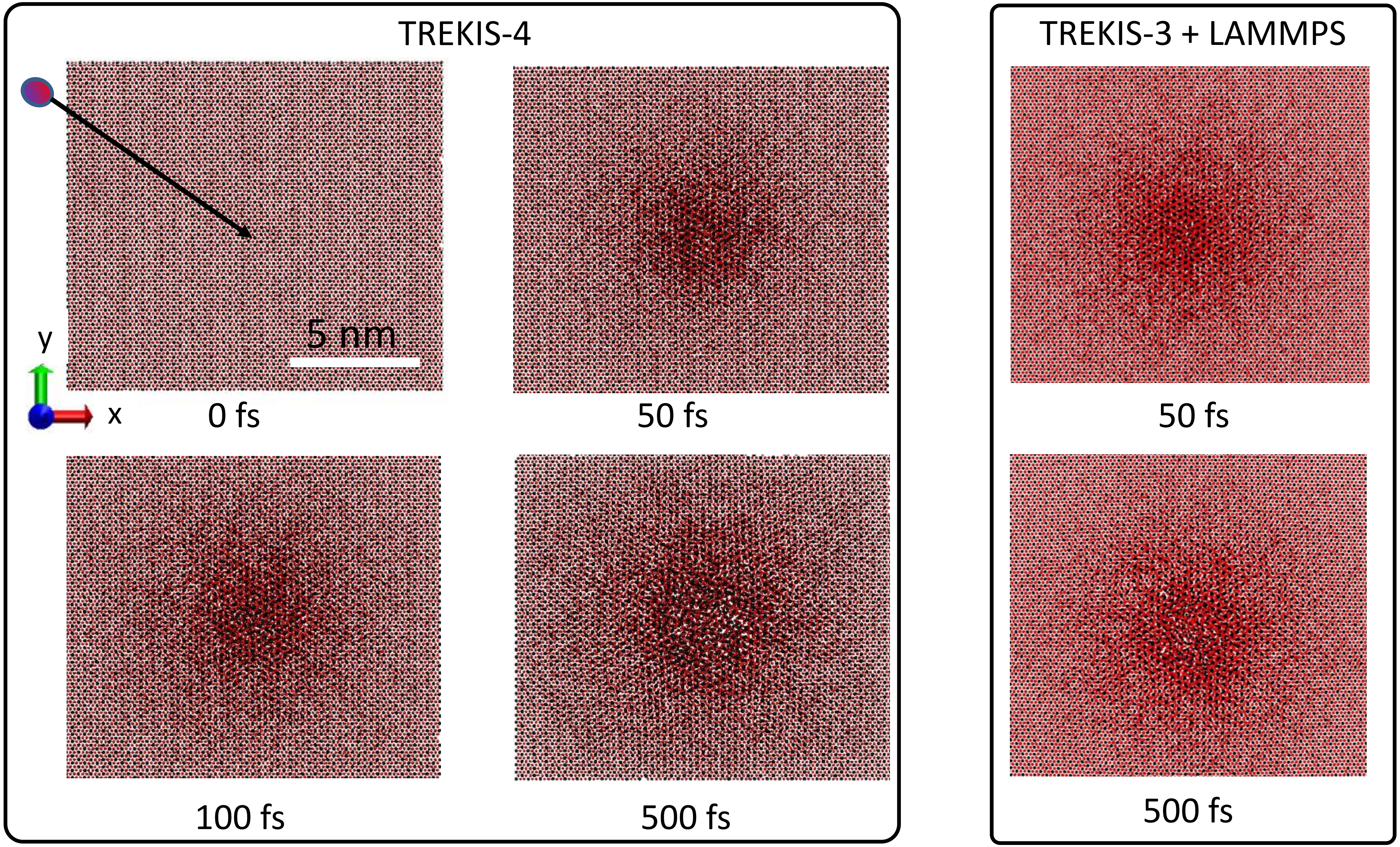
https://github.com/N-Medvedev/TREKIS-4

Example: ion beam

Medvedev *et al.*, Adv. Theory Simul. 5, 2200091 (2022)



Ion beams: with vs. without feedback

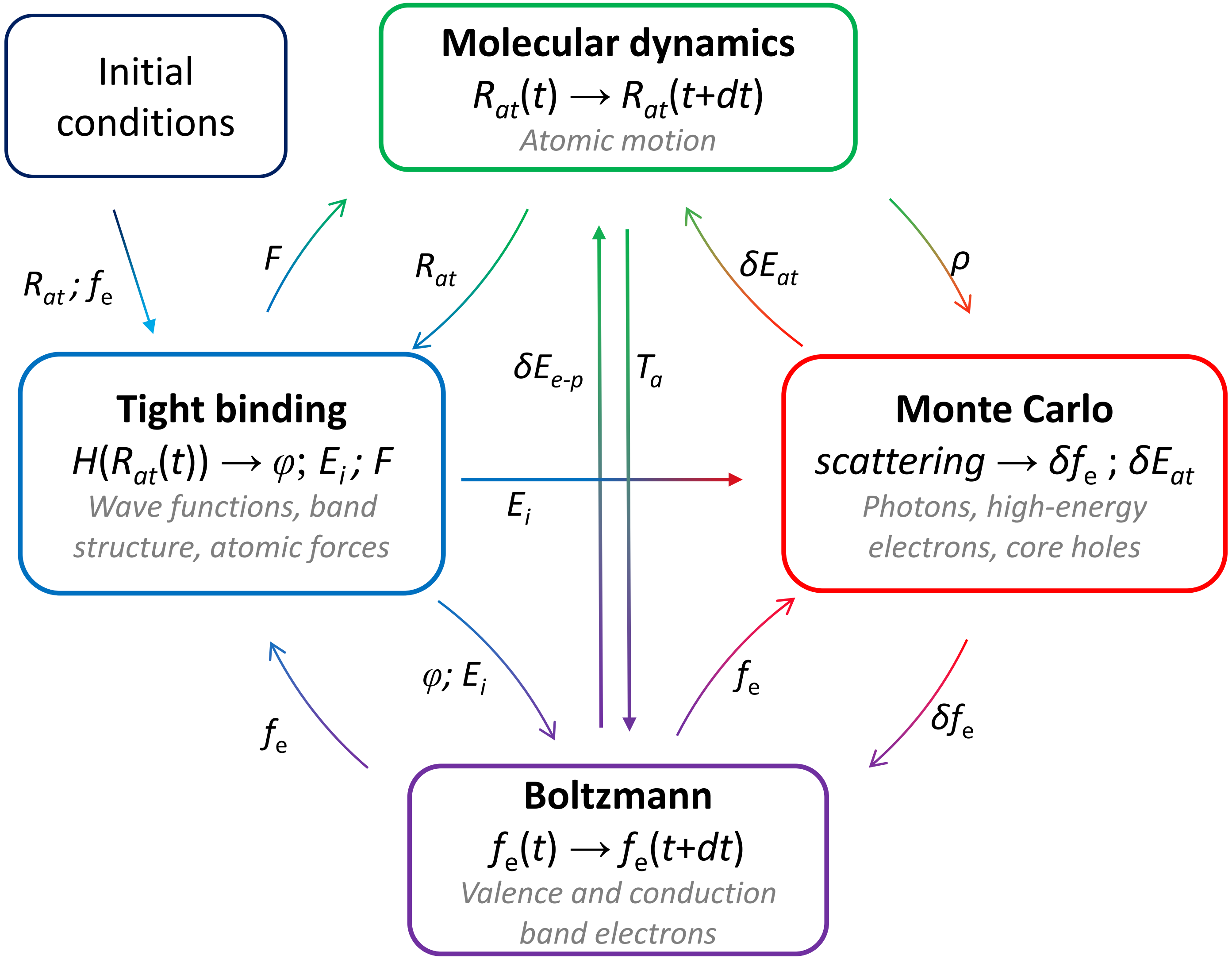


Medvedev *et al.*, Adv. Theory Simul. 5, 2200091 (2022)

Example: X-ray laser pulse

XTANT-3

X-ray-induced Thermal And Nonthermal Transitions



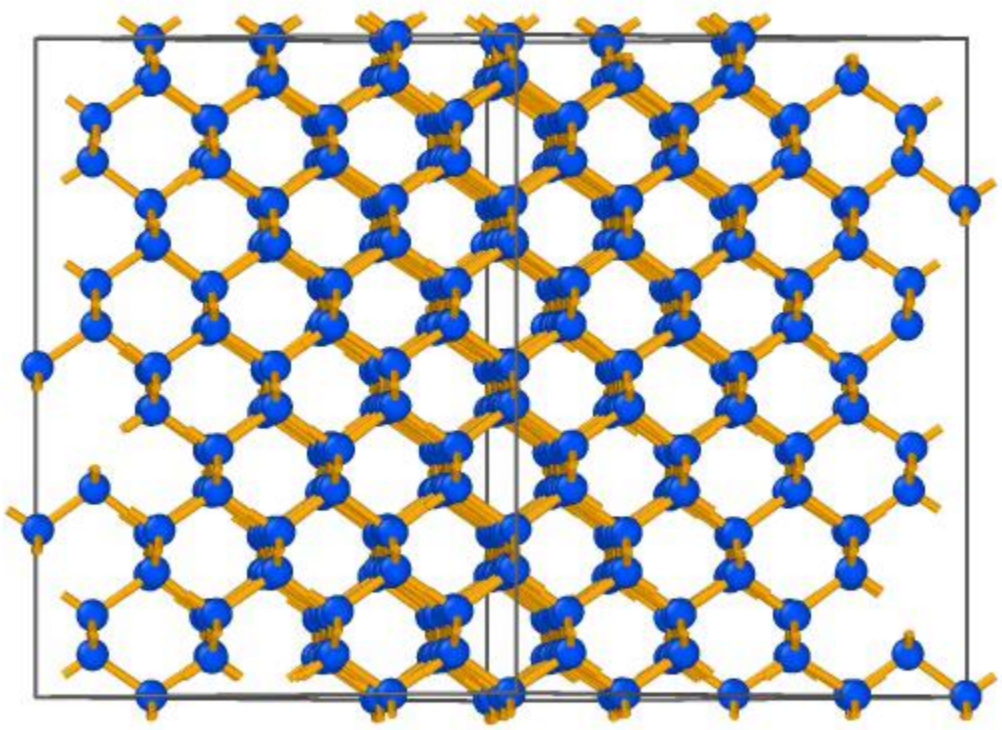
Includes :

- photon absorption
- excitation of electrons
- electron kinetics
- Auger-cascades of core holes
- equilibration of the electronic ensemble
- electron-ion (phonon) coupling
- evolution of band structure
- changes in the interatomic potential
- nonthermal melting
- atomic dynamics
- possible cooling (via thermostats)

https://github.com/N-Medvedev/XTANT-3

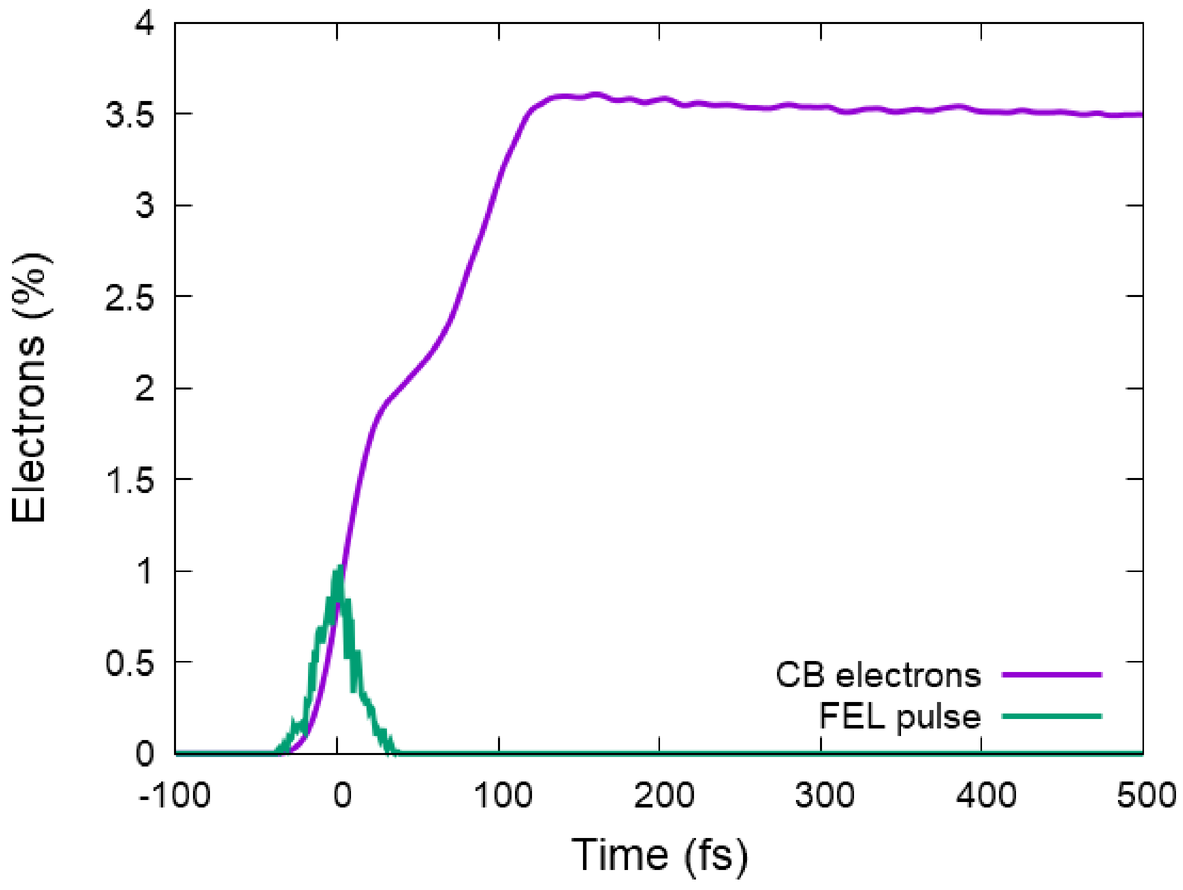
Example: X-ray laser pulse

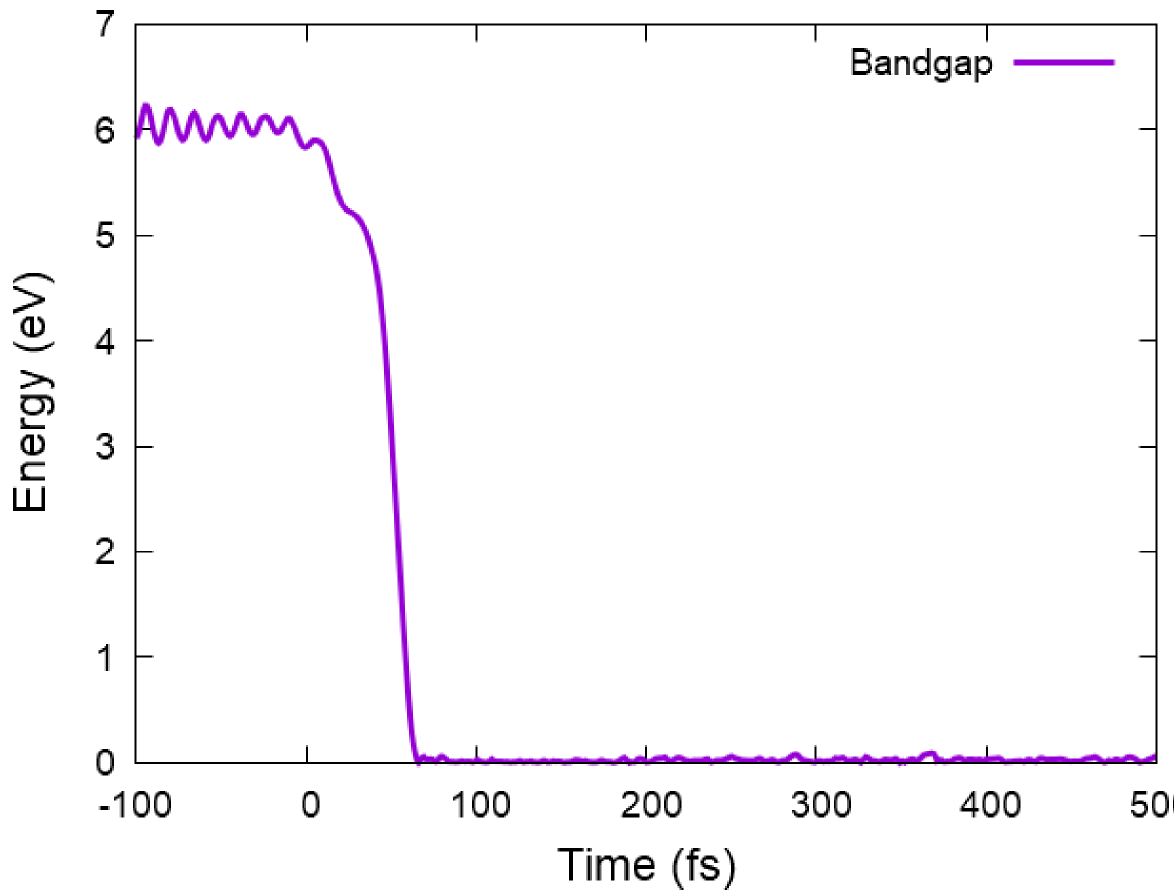
Diamond graphitization: irradiated with
10 fs pulse, 92 eV photons, 2 e V/atom dose



https://github.com/N-Medvedev/XTANT-3

Example: X-ray laser pulse



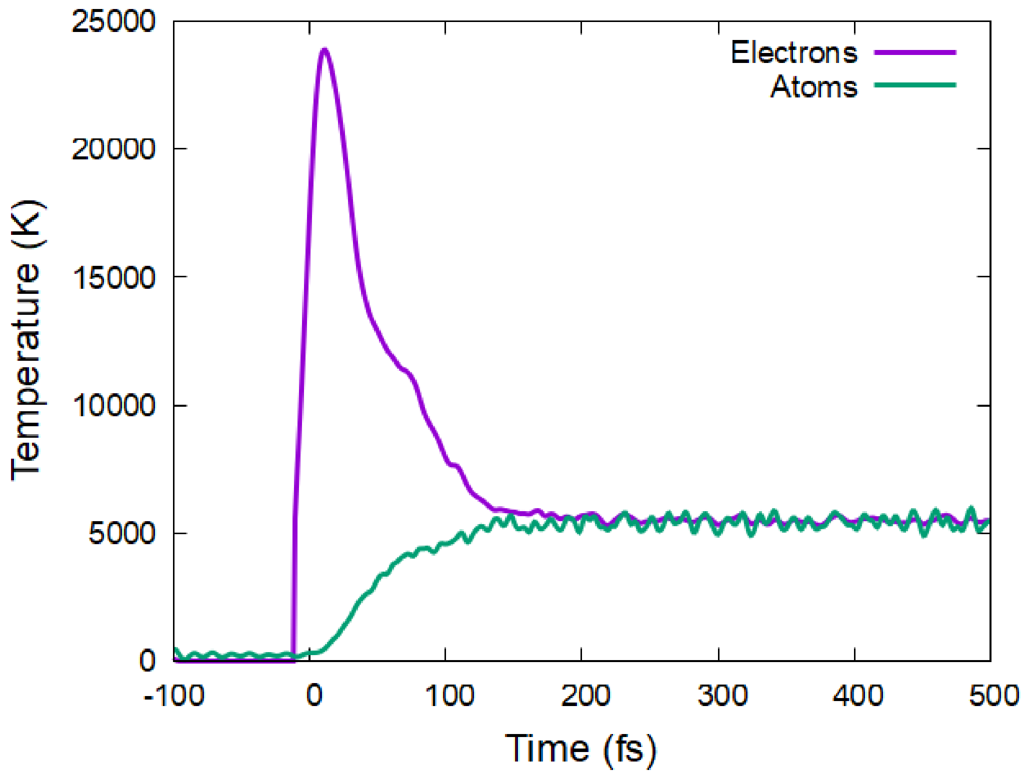


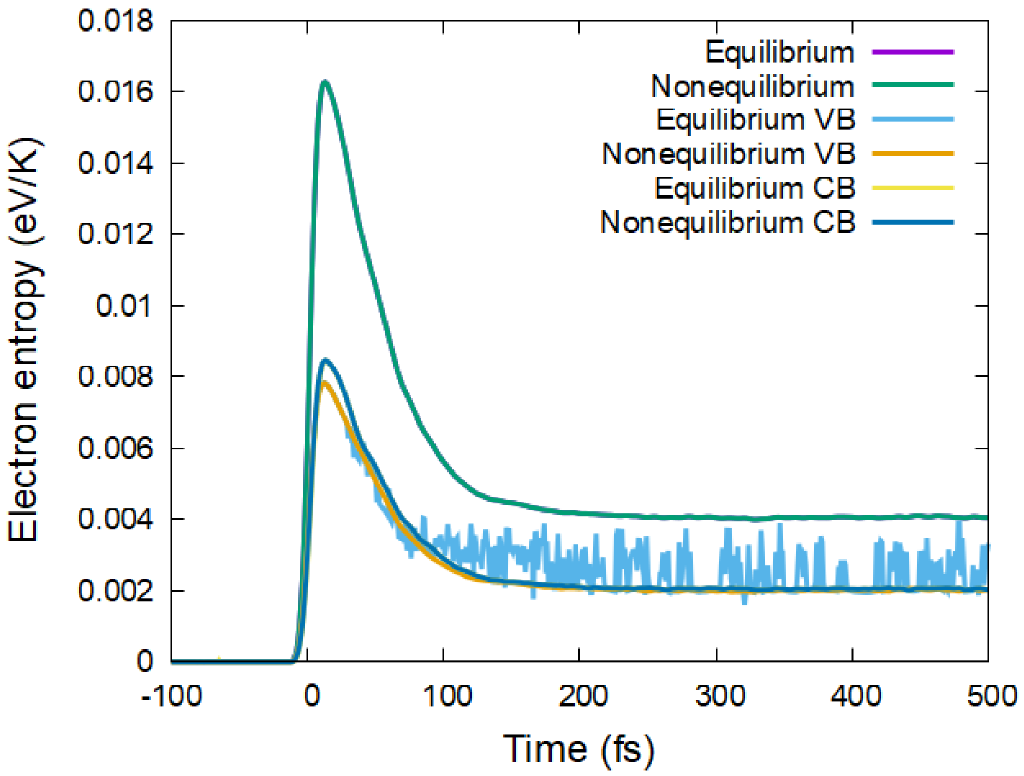
Electron density ~ 1.5 % (dose ~ 0.7 eV/atom) => phase transition

Bandgap collapse induces ultrafast phase transition

N. Medvedev *et al.*, 4open **1**, 3 (2018)

Example: X-ray laser pulse

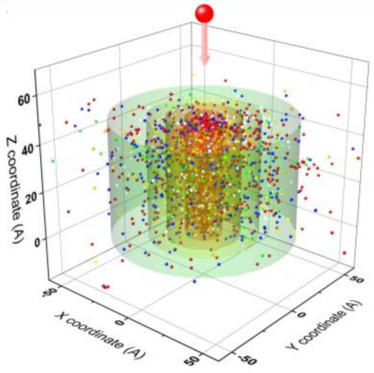




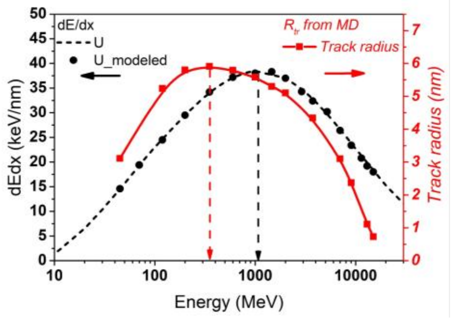
Simultaneous kinetics of electrons and atoms, not possible to model without feedbacks!

N. Medvedev *et al.*, 4open **1**, 3 (2018)

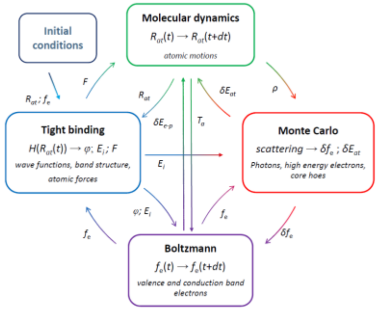
To sum up…



Transport Monte Carlo method describes evolution of the dose distribution



But! It is not sufficient to describe material damage



MC needs to be combined with other methods, describing material response (classical MD, *ab-initio* MD, etc.)

Hybrid models with feedback are needed;
models without feedbacks occasionally work too…

Institute of Physics of the Czech Academy of Sciences
FZU

Our codes

MAMBA

TREKIS3

Time-Resolved Electron Kinetics in SHI-Irradiated Solids
https://github.com/N-Medvedev/TREKIS-3

TREKIS4

Time-REsolved Kinetics in Irradiated Solids
https://github.com/N-Medvedev/TREKIS-4

XTANT3

X-ray-induced Thermal And Nonthermal Transitions
https://github.com/N-Medvedev/XTANT-3

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IPP INSTITUTE OF PLASMA PHYSICS OF THE CZECH ACADEMY OF SCIENCES