The Relaxation Cascade in Driven High-Energy-Density Matter

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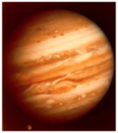
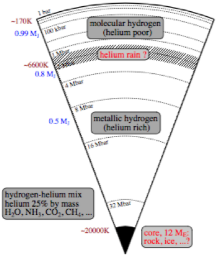
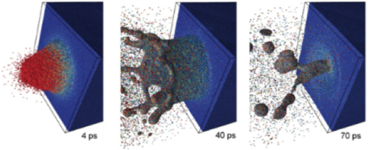
High-Energy-Density Matter: Systems of Interest

Giant planets

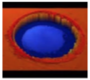




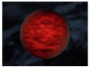
Laser ablation

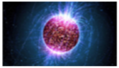


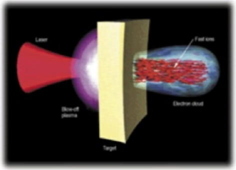
Material structuring



Colder stars

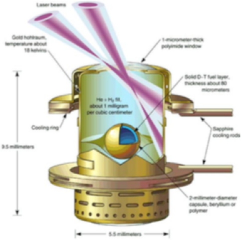






Laser ions (inside foil)

Inertial fusion energy



Why is Nonequilibrium Physics Important?

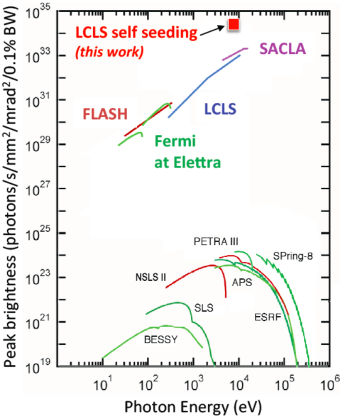
Examples for strongly driven matter

- Applications like laser ablation, nano-structuring of materials and fusion
- Short-pulse laser interactions
- Plasmas near bright stars
- Matter under FEL radiation
  - High photon numbers
  - Extreme brightness
  - Pulse duration in the fs-range
  - High repetition rate

  ⇒ FEL beam will excite matter and create a nonequilibrium state

  ⇒ FEL beam can be used to probe the equilibration process
- Some properties are highlighted in nonequilibrium situations (collisionality)

Ultra-short Pulses for Probing



from Fletcher et al., Nat. Phot. (2015)

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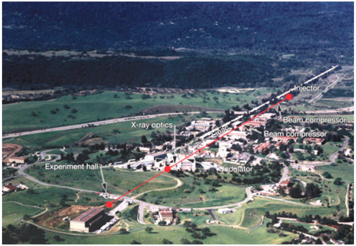


+

SACLA, European XFEL

Nonequilibrium physics of WDM becomes possible!

Outline: Relaxation Processes in Dense Plasmas

Establishment of Fermi distribution for the electrons (few fs)

$\Downarrow$

Ionisation kinetics (100s fs – few ps)

$\Downarrow$

Equilibration of ion arrangement & temperature (few ps)

$\Downarrow$

Electron-ion temperature relaxation (10s-100s ps)

$\Downarrow$

Pressure equilibration with hydrodynamic expansion
(quasi-equilibrium: motion on ns time scale)

Electron Dynamics

Kinetic Description of Electron Dynamics I

general kinetic equation within local approximation

$$\left(\frac{\partial}{\partial t} + \nabla_{\mathbf{p}} E(\mathbf{p}, \mathbf{R}t)\nabla_{\mathbf{R}} - \nabla_{\mathbf{R}} E(\mathbf{p}, \mathbf{R}t)\nabla_{\mathbf{p}}\right) f_e(\mathbf{p}, \mathbf{R}t) = \sum_b I_{eb}(\mathbf{p})$$

Weakly coupled plasmas

- Neclect collisions: $I_{eb} = 0$

⇒ Vlasov equation

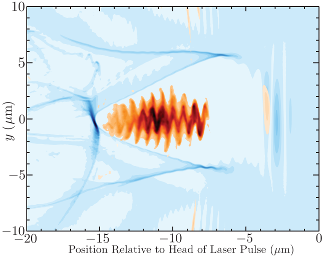
- Direct numerical solution possible
- PIC is another approach

⇒ Approaches allow for spatial and temporal dynamics

- Problem: inclusion of collisions

!!! Simple approaches $I_{eb} \sim \nu_{eb}\delta f_e$ break f-sum rule (particle conservation)

Example EPOCH output



Arber et al. using EPOCH

Kinetic Description of Electron Dynamics II

general kinetic equation within local approximation

$$\left(\frac{\partial}{\partial t} + \nabla_{\mathbf{p}} E(\mathbf{p}, \mathbf{R}t)\nabla_{\mathbf{R}} - \nabla_{\mathbf{R}} E(\mathbf{p}, \mathbf{R}t)\nabla_{\mathbf{p}}\right) f_e(\mathbf{p}, \mathbf{R}t) = \sum_b I_{eb}(\mathbf{p})$$

Strongly collisional plasmas

- Keep the collisions term I_{eb}
- Different approximations available (Boltzmann, Lenard-Balescu, ...)

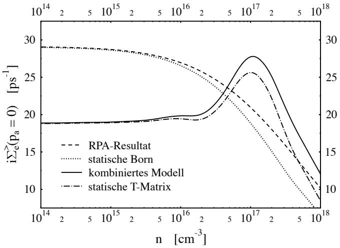
$\Rightarrow$ No solution with full drift

$\frac{\partial}{\partial t} f_e = \sum_b I_{eb}$ is often solved

$\Rightarrow$ Approach allows for temporal dynamics but for homogeneous systems only

- No problem with f-sum rule!
- Questions: can we combine with PIC?

Different collision rates



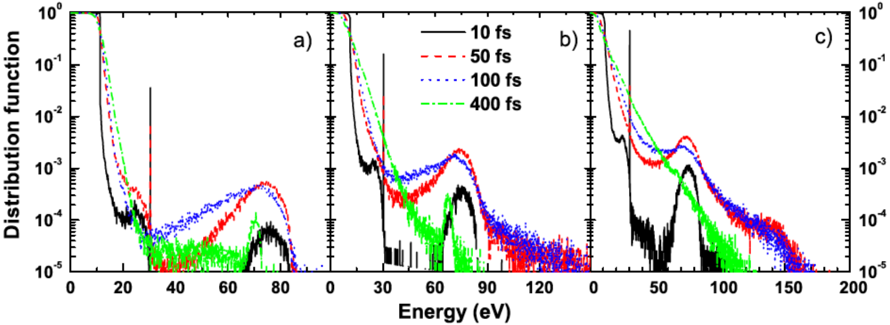
Gericke et al., PRB (1999)

Example: Electron Dynamics in FEL Excited Matter

- Electron dynamics is describe via Monte Carlo scheme
- Initial, cold distribution of the three conduction electrons in Al

⇒ Bumps at high energies due to photo-ionisation and Auger processes
⇒ Bumps quickly decay into a hot tail due to collisions
⇒ Much slower relaxation towards a full equilibrium distribution

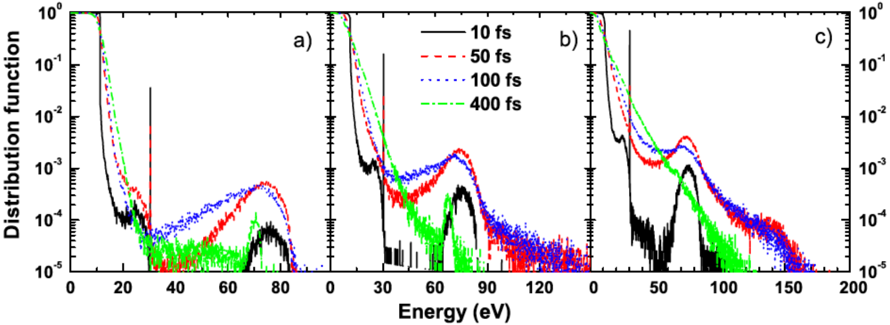
Electron distribution in aluminium pumped by VUV photons at FLASH



Electron distribution for XUV fluences of a) $0.2\,\mathrm{J/cm^2}$, b) $1.5\,\mathrm{J/cm^2}$, c) $5\,\mathrm{J/cm^2}$

Medvedev et al., PRL (2011)

Example: Electron Dynamics in FEL Excited Matter



Electron distribution for XUV fluences of a) 0.2 J/cm^2, b) 1.5 J/cm^2, c) 5 J/cm^2.

Medvedev et al., PRL (2011)

Insights for interpreting contradicting measurements

- Slow and weak heating of the conduction electron distribution

⇒ Low "temperature" (few eV) of this part explains emission spectra

- Persistent tail of high-energy electrons exists very long

⇒ Hot part of distribution (20 eV) can explain spectrum of Bremsstrahlung

Possible Test of Relaxation with X-ray Scattering

- Scattered spectrum is proportional to structure factor $S_{ee}(k,\omega)$
- For large k, scattering spectrum shows directly the distribution f_e
- In equilibrium, $S_{ee}(k,\omega)$ is obtained from response function (FDT)

$$S_{ee}(k,\omega) = \frac{\hbar}{\pi n_e} \frac{1}{1-\exp(-\beta_e \hbar\omega)} \, \mathrm{Im}\chi_{ee}(k,\omega)$$

- How do we calculate $S_{ee}(k,\omega)$ in nonequilibrium situations?
- Extension of fluctuation-dissipation theorem in nonequilibrium

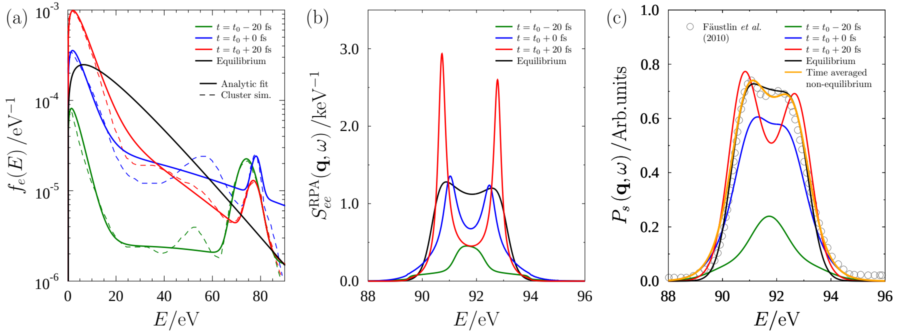
$$S_{ee}(k,\omega) = \frac{i\hbar}{2\pi n_e} \frac{\Pi^{>}_{ee}(k,\omega)}{|\varepsilon(k,\omega)|^2} \overset{\text{RPA}}{=} \frac{S^0_{ee}(k,\omega)}{|\varepsilon(k,\omega)|^2}$$

- Ideal structure factor given by distribution functions
- Screening function in RPA is also given by distribution functions

$\Rightarrow$ Mode spectrum is modified for nonequilibrium situations

Example: Predictions for VUV Self-Scattering

Analysis of FLASH experiment using nonequilibrium FDT



Chapman & Gericke, PRL (2011)

- Analysis reveals a very dynamic behaviour of the scattering spectrum
- Plasma parameters strongly evolve und differ from equilibrium fit
- Nonequilibrium analysis yield good agreement with experiments

$\Rightarrow$ **Nonequilibrium dynamics can, in principle, being tested!**

Ionisation Kinetics

Concepts for Ionisation Equilibrium in Dense Matter

Plasma Physics Concept

- Start from electronic structure of isolated atom
- Saha equation determines the charge state distribution (equilibrium)

$$\frac{n_i}{n_{i+1}} = \frac{g_i}{g_{i+1}} \exp(\beta\mu) \ \exp(\beta E_i^{\mathrm{eff}})$$

- Relaxation via system of rate equations with $\alpha(E_i^{\mathrm{eff}})$ and $\beta(E_i^{\mathrm{eff}})$
- Introduce effective ionisation energies $E_i^{\mathrm{eff}} = E_i^0 + \Delta_i$
- Shift $\Delta_i(n, T, Z_i)$ accounts for interactions with surrounding medium

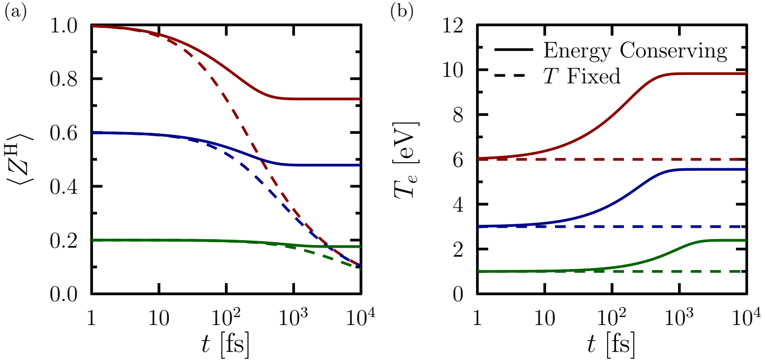
Solid State Physics Concept

- Assume fixed configuration of nuclear charges
- Calculate band structure for the electronic states
  (valence band: 'bound'; conduction band: 'free')
- Insure that ionic configuration is consistent with electronic structure

Example: Evolution of Ionisation and Temperature

- Consider hydrogen plasma with $n_p = 10^{21}\,\mathrm{cm}^{-3}$
- 3 different initial ionisation levels
- Consider either fixed electron temperature or coupled evolution

⇒ Solve coupled rate equations for charge states and temperature



Baggott (PhD thesis)

Results strongly depend on effective ionisation energy (IPD) !

Recent Experimental Results for the IPD

LCLS	ORION	NIF
• Aluminium • 70-180 eV • 2.7 g/cm^{-3}	• Aluminium • 550-700 eV • 1.2-9.0 g/cm^{-3}	• Plastics / C • 50-200 eV • 4.0-20.0 g/cm^{-3}
• Matched by Ecker-Kröll • Not matched by Stewart-Pyatt • Not matched by Debye	• Not matched by Ecker-Kröll • Matched by Stewart-Pyatt • Not matched by Debye	• Not matched by Ecker-Kröll • Not matched by Stewart-Pyatt • Matched by Debye

These are quite diverse results/parameters ... what can we learn?

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These are quite diverse results/parameters ... what can we learn?

Theories based on nonlinear screening provide (some) explanation

IPD with Nonthermal Electrons: Theory

- Start with linear response of electrons to ionic fields
- Change in ionisation energy is still $\Delta = -\kappa e^2$
  (assumes calculated fields at origin where ion is located)
- Dielectric function in RPA yields dynamic screening
- Take static limit and then the long wavelength limit
  (order is important)
- Static screening length in nonequilibrium is obtained as

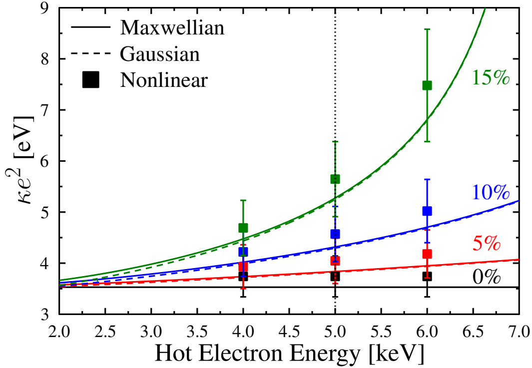
$$\kappa^2(t) = \frac{e^2}{\varepsilon_0} m_e \, 4\pi \int_0^\infty \frac{dp}{(2\pi\hbar)^3} \, f_e(p,t)$$

$\Rightarrow$ Low sensitivity to hot electron distribution function

- Extension with nonlinear screening and extended ions are possible

IPD with Nonthermal Electrons: Results

Consider carbon under X-ray free electron laser conditions (LCLS)



Baggott & Gericke (tbp)

Dashed line indicates 'bump' feature found in MD simulations
Hau-Riege, PRE (2013)

- Hot electrons contribute little to the screening and IPD
- Hot electrons act as an energy sink keeping bulk electrons colder
- Increased screening in systems with hot electrons for fixed energy

Build-Up of Ionic Correlations

Build-up of Ionic Correlations & Structure

Models for a theoretical description

- Direct (classical) molecular dynamics simulation in new energy landscape
  Murillo, PRL (2001)
- Final state from energy conservation applying the new pair distribution
  Gericke *et al.*, J.Phys.A (2003)

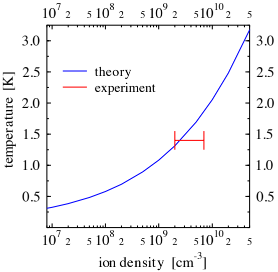
Extreme example: ultra-cold plasmas

- No initial correlations (ideal gas)
- Almost no kinetic energy at $t=0$ (gas temperature $\approx 1\,\mu$K)

$\Rightarrow$ $U_{ii}^{corr}(0)=0$ and $E_i^{kin}(0)=0$

$\Rightarrow$ Effective coupling strength:

$$\Gamma_{ii}^{eff}(t)=\frac{\left|U_{ii}^{corr}(t)\right|}{E_i^{kin}(t)}=\frac{\left|U_{ii}^{corr}(t)\right|}{E_i^{kin}(0)+\left|U_{ii}^{corr}(t)\right|}\rightarrow 1$$



exp.: Killian et al., PRL (2005)

Other important example: ultra-fast nonthermal melting

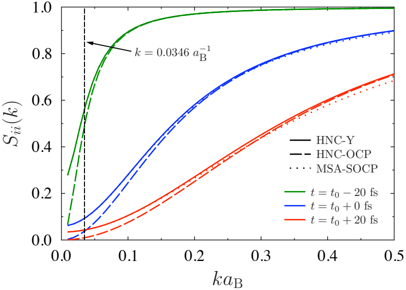
Example: “Missing” Elastic Peak in XRTS Data

Full spectrum should contain an elastic scattering peak

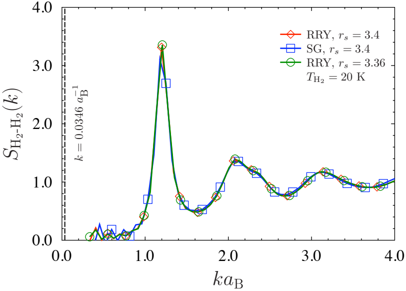
$$P(\theta,\omega) \sim S_{ee}^{tot}(k,\omega) = |f_i(k) + q(k)|^2 \, S_{ii}(k)\delta(\omega) + Z_f \, S_{ee}^0(k,\omega)$$

⇒ **What is the static ion structure in the system at probe time?**

Coulomb interactions (T=20 K)



Original hydrogen liquid



Chapman et al., HEDP (2012)

⇒ Ions retain their structure from the cold liquid during the pulse

⇒ **The ion-ion structure factor for the k-value probed is very small**

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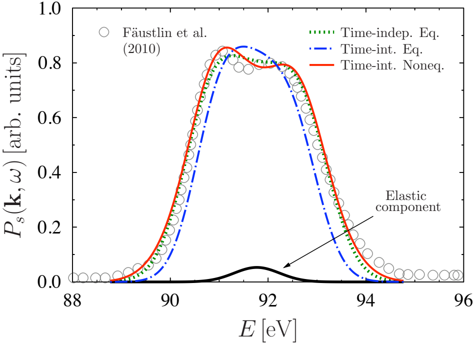
Applying initial hydrogen structure to scattering spectrum



Full power spectrum for FEL driven hydrogen (self-scattering)

- **Excellent agreement with experimental data**
- Best fit with data for:
  $S_{ii}(k_{\mathrm{probed}}) = 0.04$
- Ionic correlations are not present on fs-time scales
- We need to consider yet another relaxation process: **Build up of Ion Correlations**

Chapman et al., HEDP (2012)

Electron-Ion Energy Relaxation

Theoretical Models for the Energy Transfer

- Landau-Spitzer approach for weak classical electron-ion collisions

$$\dot{T}_e = (T_i - T_e)\frac{8\sqrt{2\pi}Z_i^2 e^4 \ln\lambda_c}{3m_e m_i}\left(\frac{T_e}{m_e} + \frac{T_i}{m_i}\right)^{-3/2}$$

Landau (1936), Spitzer (1967)

- Strong binary collision within quantum kinetic theory

$$E_{e\to i}^{trans} = \frac{1}{2\pi\hbar^3}\frac{n_e\Lambda_e^3}{m_i m_r}\int_0^\infty dk\, k^5\, Q^T(k)\, \exp\left(-\frac{k^2}{2m_e k_B T_e}\right)$$

Gericke *et al.*, PRE (2002)

- Energy transfer through coupled collective modes

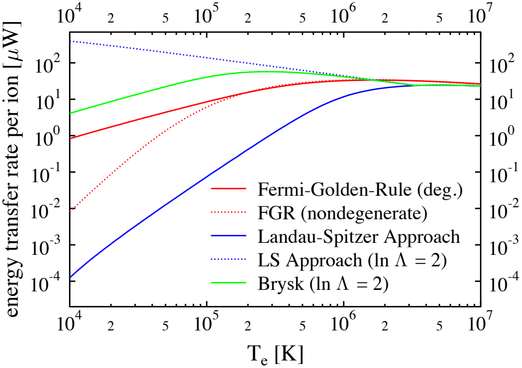
$$E_{e\to i}^{trans} = 4\hbar\int_0^\infty \frac{d\omega}{2\pi}\,\omega\int\frac{d^3\mathbf{k}}{(2\pi)^3}\left|U_{ei}^S(k)\right|^2\frac{\Delta N_{ei}\,\chi_e''(\omega,k)\chi_i''(\omega,k)}{1 - V_{ei}(k)\chi_e(\omega,k)\chi_i(\omega,k)}$$

Dharma-wardana & Perrot, PRE (1998)

- Fermi's-Golden-Rule approach (simplest model with collective modes)

Example: Electron-Ion Energy Transfer

Results for the energy transfer rates



Energy transfer rates for silicon plasmas with $Z_i = 4$, $n_i = 1.17 \times 10^{23}\,\mathrm{cm}^{-3}$, and $T_i = 10^3\,\mathrm{K}$. Parameters like Celliers et al., PRL (1992)

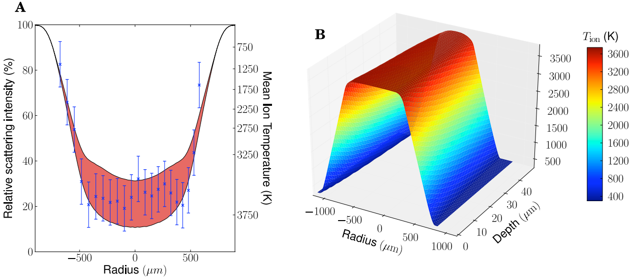
Insights gained

- LS approach fails for degenerate plasmas
- Brysk describe rates only qualitatively
- What about coupled mode effects?
- **Huge theoretical uncertainties!**

Measuring Electron-Phonon Coupling in Graphite

- Experimental setup: Proton-heated sample probed by x-ray diffraction
- Protons mainly heat electrons and set initial conditions $T_e(0)$
- Lattice temperature is measured via decrease in Bragg scattering
- DFT-MD provides EOS model and Debye temperature/Debye-Waller factor

at $t = 225$ ps



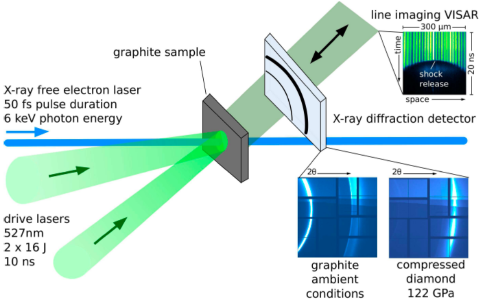
- Changes in lattice temperature can only explained by an extremely low electron-phonon constant of $g = 4.5 - 8 \times 10^{15}\,\mathrm{WK^{-1}m^{-3}}$.

$\Rightarrow$ **Strong evidence of energy transfer bottleneck in WDM**

White et al., Scientific Reports (2012)

Material Modifications

Example: Modifying Materials with High Pressures

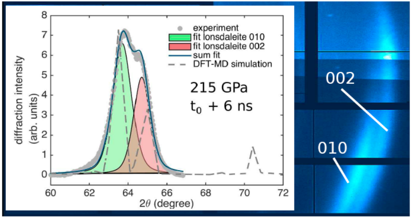


- Amorphous graphite sample
  ⇒ Debye-Scherrer rings
- Position of DS-rings indicate compression
- Very clear signal with FEL
- Ultra-fast probing possible

⇒ Evolution of phase transition

Theory

- rings need to be identified with lattice structure
- occurrence of new rings
  ⇒ structure search needed
- here: position consistent with lonsdaleite



Kraus et al., Nature Communications (2016)

... as a Summary

momentum relaxation electrons: $\sim 1\,$**fs**

ionisation equilibrium; rate equations: $\sim 1\,$**ps**

momentum relaxation ions: $\sim 100\,$**fs**

temperature relaxation $\sim 10\,$**ps**

Transient processes offer a window to rich & interesting physics and FELs, combined with high-energy laser, are a perfect tool to investigated the different relaxation stages toward equilibrium.

One has to resist the "equilibrium trap" when analysing the data!

Thank you!