



Laser Plasma Interaction

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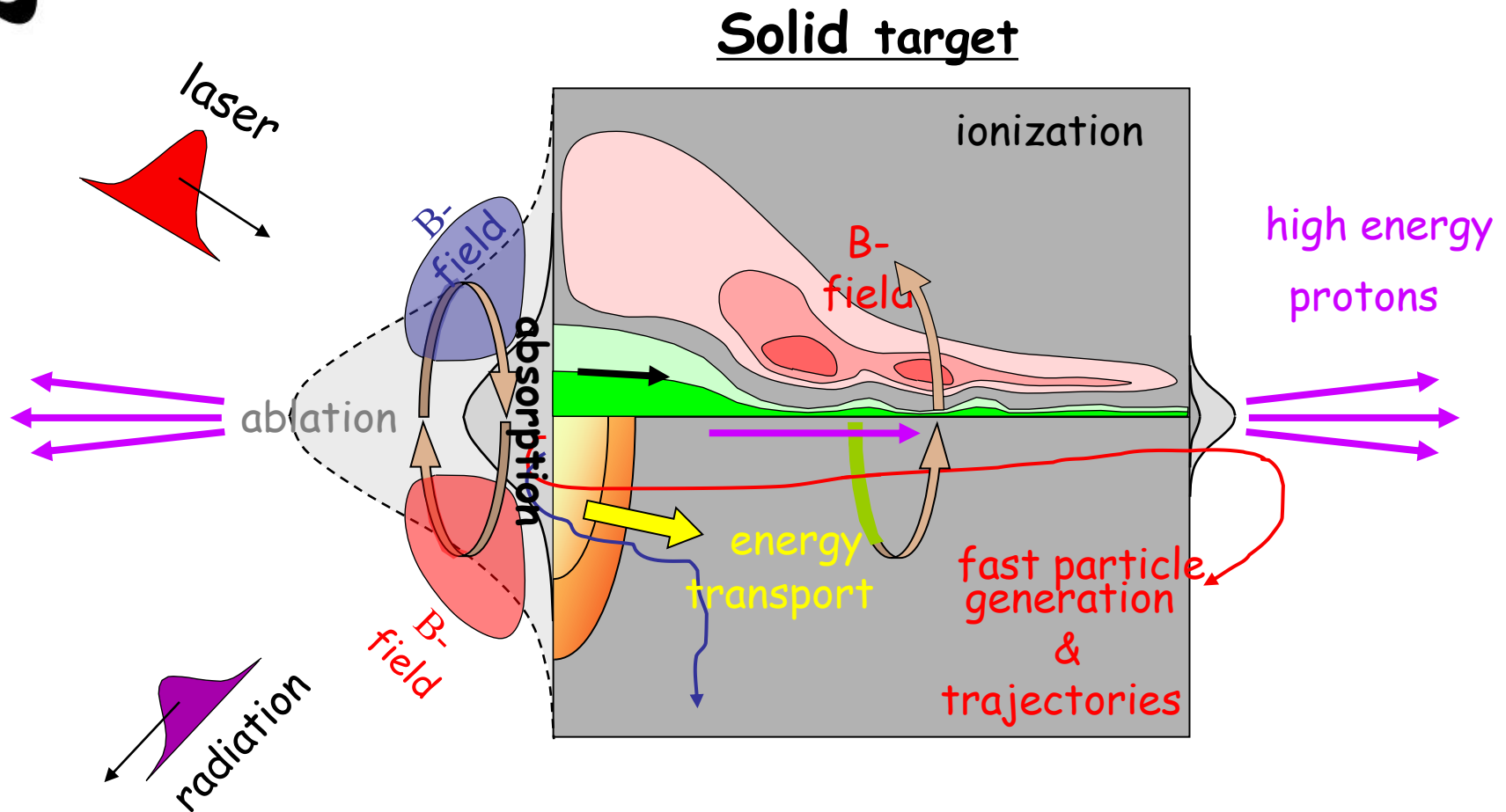
Outline

- **Introduction.**
 - What is plasma?
 - What is Laser?
 - Laser Ionization dynamics.
 - ns versus fs lasers (Plasma parameters).

- **Applications**
 - Laser induced breakdown spectroscopy (LIBS).
 - Plasma deposition and etching.
 - UV and x-ray production.
 - Warm dense matter.



Laser-Matter Interaction

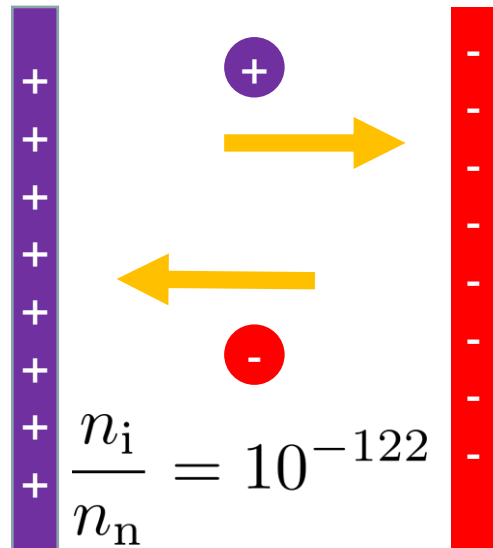


Short pulse laser: Interaction with matter, P. Gibbon, 2005

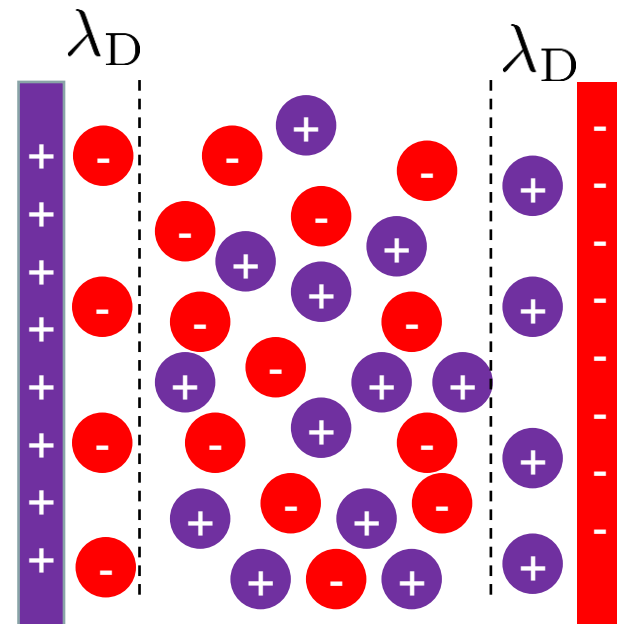


What is plasma?

- The plasma is a quasineutral gas of charged and neutral particles which exhibits collective behavior.



Air at room temp.



PLASMA

Somthing molded or fabricated



What is Laser?

- The laser is a beam of photons characterized with :
Monochromatic, Directionality, Coherent, High intensity.

- *Conventional Lasers:*

- *Active medium*
- *Pumping*
- *Population inversion*
- *Stimulated emission*
- *Resonator*



Wicked Lasers

- ***X-ray Free electron Lasers !!!!***



Light matter interaction

- Photoelectric effect: Ionization takes place if the photon energy greater than the work function of the matter.

$$\hbar\omega_{\text{photon}} = W_{\text{function}} + E_{\text{kin}}$$

→
 $\hbar\omega_{\text{photon}}$

W_{function}



- *A. Einstein: Nobel Prize 1921*

- *Ti:Sa Laser 800 nm with photon energy $\hbar\omega = 1.5\text{eV}$*

Material	Cs	Li	Na	Hg	Au
W. function	2.1 eV	2.9 eV	2.3 eV	4.5 eV	5.5



Laser matter interaction I

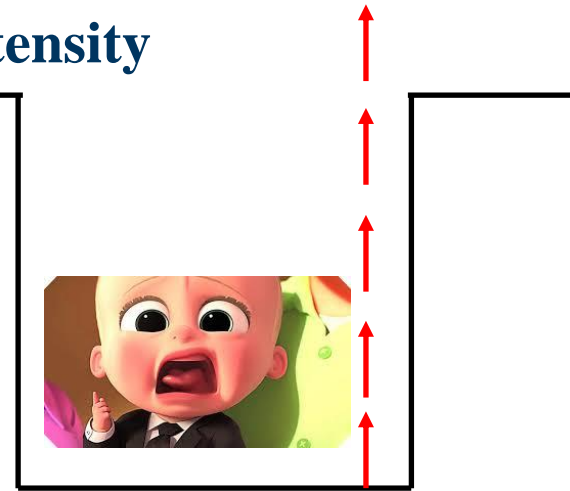
- Multiple Photon ionization : one single atom can interact with multiple photons at the same time.
- The rate of ionization depends on the laser intensity

$$\Gamma^n = \sigma_n I_L^n$$

$$n\hbar\omega$$

- **Ti:Sa Laser with intensity**

$$10^{12} W/cm^2 \approx 10^{31} \text{ photon}/cm^2 \text{ Sec}$$



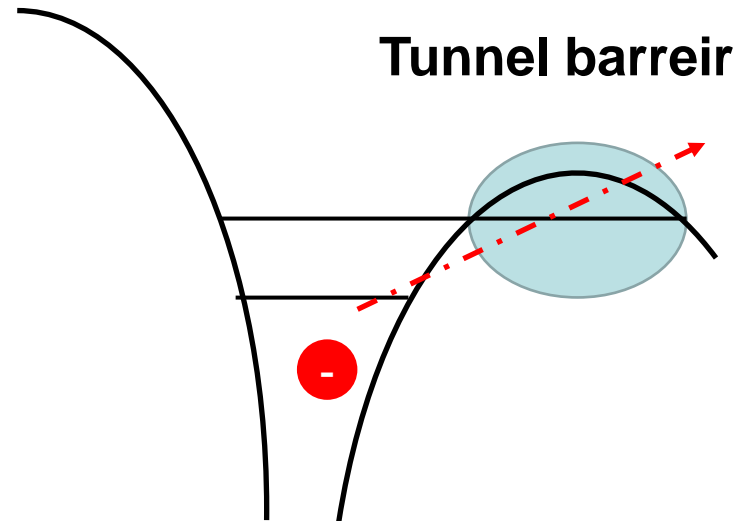
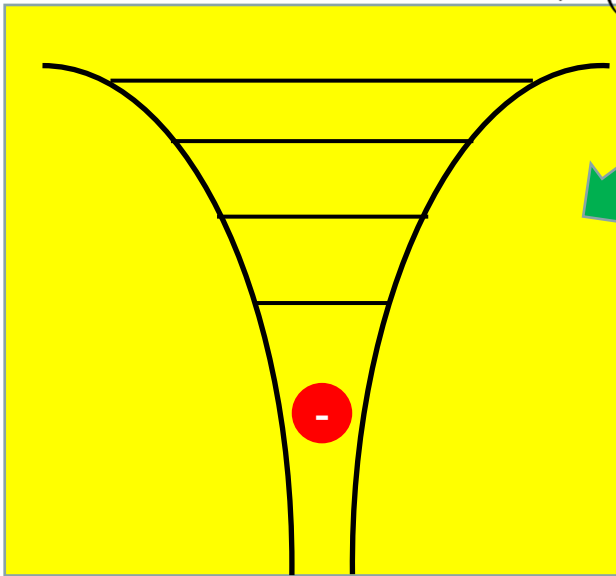
Material	Ionization energy	N- photons
H ₂	13.6 eV	9 photons
He	24.5 eV	16 photons



Laser matter interaction II

- Tunnel ionization: The potential of the laser field can modify the Coulomb potential of an electron in an atom and forms a potential barrier. It depends on the barrier width and height.

$$V(r, t) = -\frac{Ze^2}{4\pi\epsilon_0 r} - eE(t)r$$





Laser matter interaction III

- **Keldysh Parameter:** depends on the ionization energy of an electron ξ to the pondermotive potential $e\Phi$.

$$\gamma = \sqrt{\xi/e\Phi} = \sqrt{\xi/(\lambda_L^2 I_L)}$$

- **Tunnel ionization** $\gamma \leq 1$ **MPI** $\gamma > 1$
- **For Ti:Sa Laser interacts with He:**

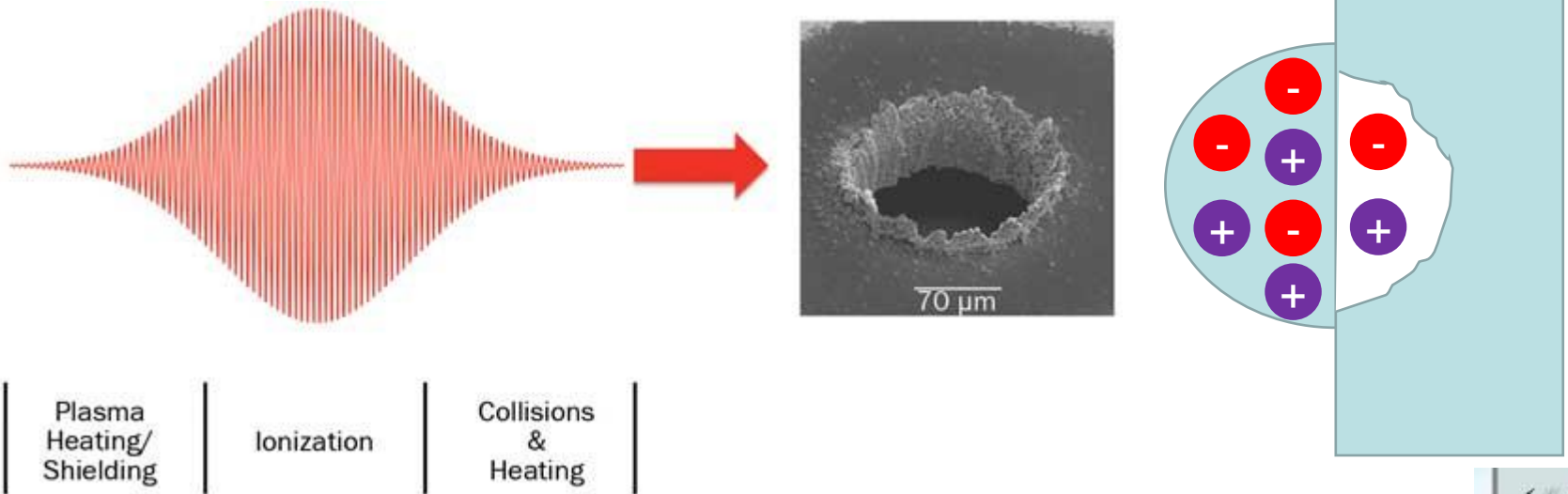
Intensity	Keldysh parameter	Ionization
10^{12} W/cm^2	14	MPI
$2 * 10^{14} \text{ W/cm}^2$	1	Tunnel

- **Barrier suppression ionization (BSI):** At higher laser intensities electrons can leave atoms without tunneling : dominant above 10^{18} W/cm^2 .



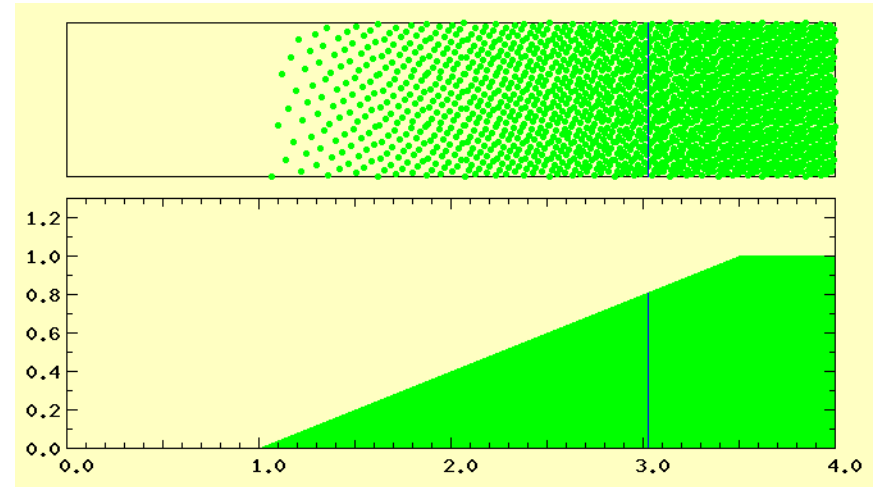
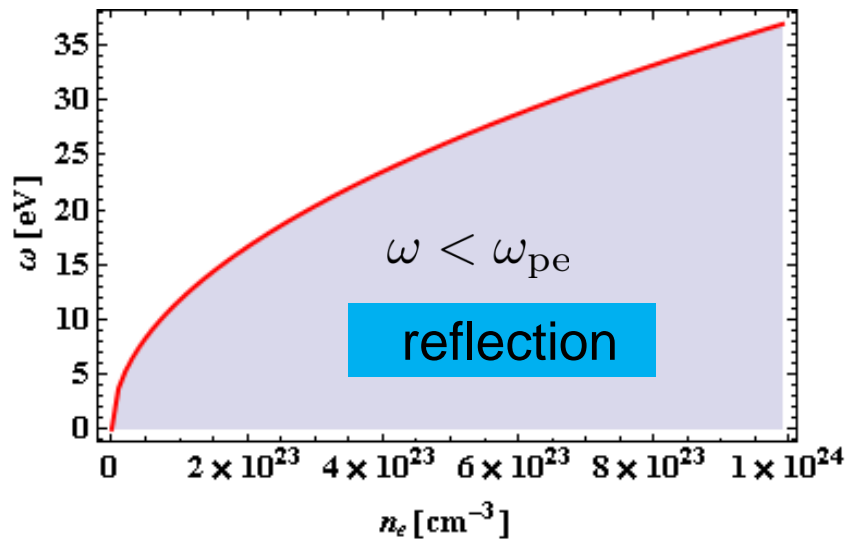
Plasma creation over solid targets

- The first seed of electrons are generated via MPI, Tunnel ionization, or , BSI.
- The free electrons gain energy from the laser electric field, then make further ionization by collision with neutral particles in the target.





EMW Dispersion relation



Warwick UK

- *The plasma refractive index*
- *The critical density*

$$n_{el}^2 = 1 - \frac{\omega_p^2}{\omega^2}$$

$$n_c = 1.1 \times 10^{21} \lambda_L^{-2} (\mu m) cm^{-3}$$



Nano-second Pulses I

- The target is heated via electron-ion collisions to 10s to 100s eV depends on the laser intensity

$$T_e \approx 13 \times 10^7 (I_{\text{abs}} (W/\text{cm}^2) / n_e)^{2/3} \text{eV}$$

- The plasma pressure created during heating causes ion blow-off (ablation) at the sound speed

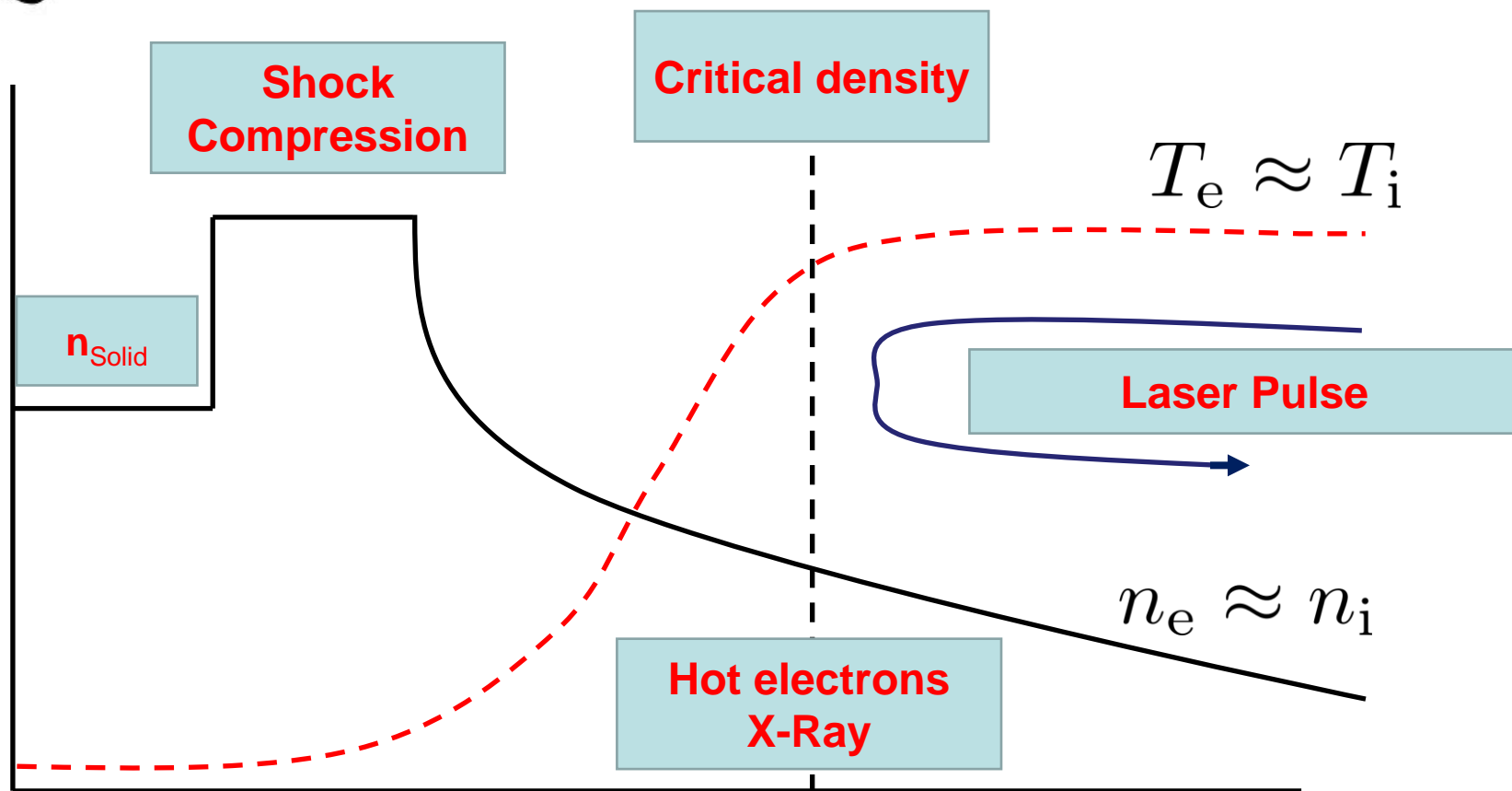
$$C_s \approx 3.1 \times 10^7 (T_{\text{KeV}} Z^*)^{1/2} \text{cm/s}$$

- Because of ablation, density profile decreases exponentially with a scale length of

$$L = C_s \tau_L$$



Nano-second Pulses II





Absorption Mechanisms I

- **Inverse Bremsstrahlung absorption:** The electrons, while oscillating under the action of the laser electric field, collide with the ions giving rise to transfer of electromagnetic energy to the plasma.

$$K_{ib} \approx 3.1 \times 10^{-7} Z n_e^2 \ln(\Lambda) \omega_L^{-2} \left[1 - \frac{\omega_p^2}{\omega_L^2} \right]^{-1/2} T_{eV}^{-3/2} \text{cm}^{-1}$$

- **The fraction of absorbed laser energy after a propagation over a distance L in a uniform plasma is**

$$\alpha_{abs} = 1 - \exp[-K_{ib}L]$$

- **Ion turbulence absorption:** Inverse Brem. Absorption increases significantly when ion motion is correlated.



Absorption Mechanisms II

- **Resonance absorption:**
 - **Laser radiation obliquely incident on a plasma and with $(\vec{E}_L \cdot \vec{\nabla}(n_e) \neq 0)$ can excite resonant longitudinal plasma oscillations at critical density surface.**
 - **The damping of the excited electron waves leads to conversion of electromagnetic laser energy into thermal energy.**

- **The amplitude of the electrons oscillations depends on the laser polarization:**

$$\delta n_e \propto \frac{\vec{E}_L \cdot \vec{\nabla}(n_e)}{n_e - n_c}$$





- **S-Polarized $(\vec{E}_L \cdot \vec{\nabla}(n_e) = 0)$ can not excite plasmons or Langmuir waves.**



Absorption Mechanisms III

- **Parametric instabilities: Wave-wave interaction**

$$\omega_0 = \omega_1 + \omega_2 \quad K_0 = K_1 + K_2$$

- **Photon**  **Photon + Acoustic**
Stimulated Brillouin scattering
- **Photon**  **Photon + Plasmon**
Stimulated Raman scattering
- **Photon**  **Acoustic + Plasmon**
Decay instability
- **Photon**  **Plasmon + Plasmon**
Tow-Plasmon instability



Femto-second Lasers

- **Brunel effect:** Intense laser pulses incident on sharp overdense plasmas, pulls electrons into the vacuum and then back them into the plasma.
- **Filamentation:** Energetic beam of electron penetrate into the core of the target producing ionization.
- **Surface wave:** Sharp overdense plasma boundary supports surface waves; electron fluctuations accompanied with electromagnetic waves.
- **And more**



Radiation Hydrodynamic

- **Governing equations in planar geometry:**
- **The independent Lagrangian mass variable: The spatial grids moves with the fluid** $dm_0 = \rho(r)dr$
- **The momentum conservation equation**

$$\frac{\partial u}{\partial t} = -\frac{\partial}{\partial m_0}(P + q)$$

- P is the total pressure due to electrons, ions, and radiation,
 q is Neumann artificial viscosity
- **Conservation of energy**

$$C_{v,e} \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial m_0} \xi_e \frac{T_e}{\partial r} - \omega_{ei}(T_e - T_i) + R_{\text{Abs}} - R_{\text{Emis}} + S_e - \dots$$

$$C_{v,i} \frac{\partial T_i}{\partial t} = \frac{\partial}{\partial m_0} \xi_e \frac{T_i}{\partial r} + \omega_{ei}(T_e - T_i) - \dots$$



Local thermodynamic equilibrium

- **Thermodynamic equilibrium:** require that plasma fulfils four energy distributions; radiative, kinetic, excitation, and ionization energy, termed, the Planck, Maxwell, Boltzmann, and Saha relations; each energy exchange process must be balanced by its inverse. (Not found in man made plasmas).
- **Local thermodynamic equilibrium;** when collisional processes are more important than radiative, one can find a temperature based on Maxwell, Boltzmann, and Saha distributions.
- **All atomic processes must be considered:** radiative ionization, radiative excitation, radiative recombination, impact ionization, dielectronic recombination, three body recombination,



Atomic physics calculations

- **Atomic rate equations:**

$$\frac{dn_i}{dt} = -n_i \sum_{i \neq j}^{N_L} W_{ij} + \sum_{i \neq j}^{N_L} n_j W_{ji}$$

- **Upward (depopulating) and downward (populating) transitions:**

$$W_{ij} = \underbrace{n_e C_{ij} + B_{ij} \overline{J_{ij}}}_{excitation} + \underbrace{n_e \gamma_{ij} + \beta_{ij} + \Omega_{ij}}_{ionization}$$

$$W_{ji} = \underbrace{n_e D_{ji} + A_{ji} + \beta \overline{J_{ij}}}_{deexcitation} + \underbrace{n_e^2 \delta_{ji} + n_e (\alpha_{ji}^{RR} + \alpha_{ji}^{DR})}_{recombination}$$

- **Continuum lowering effects are considered**



ns versus fs pulses I

- **Two laser pulses**

- 1ns, 0.015 TW/cm^{-2}

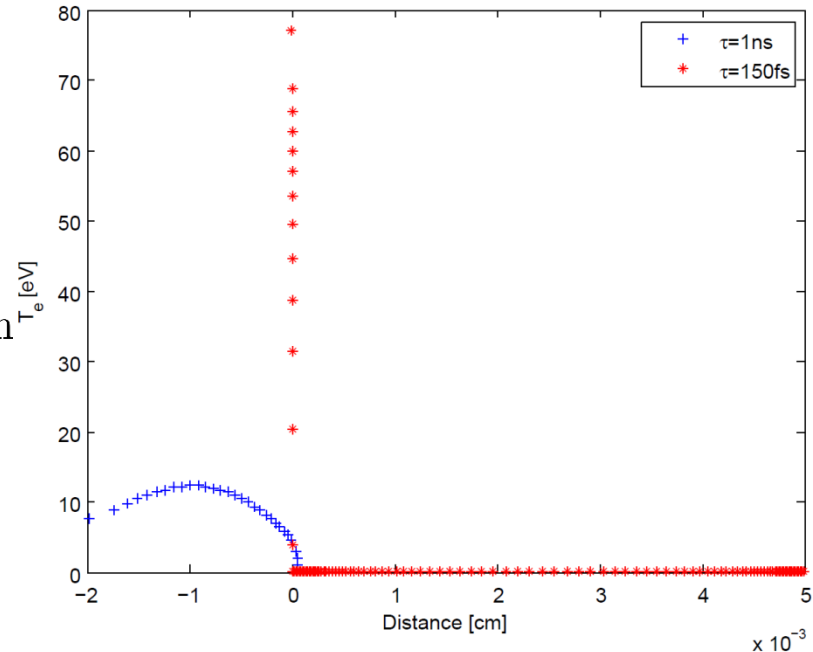
- 150fs, 100 TW/cm^{-2}

- **Ar droplet with diameter** $50 \mu\text{m}$

- **Wavelength of** 810 nm .

- **Energy of** 0.023 mJ .

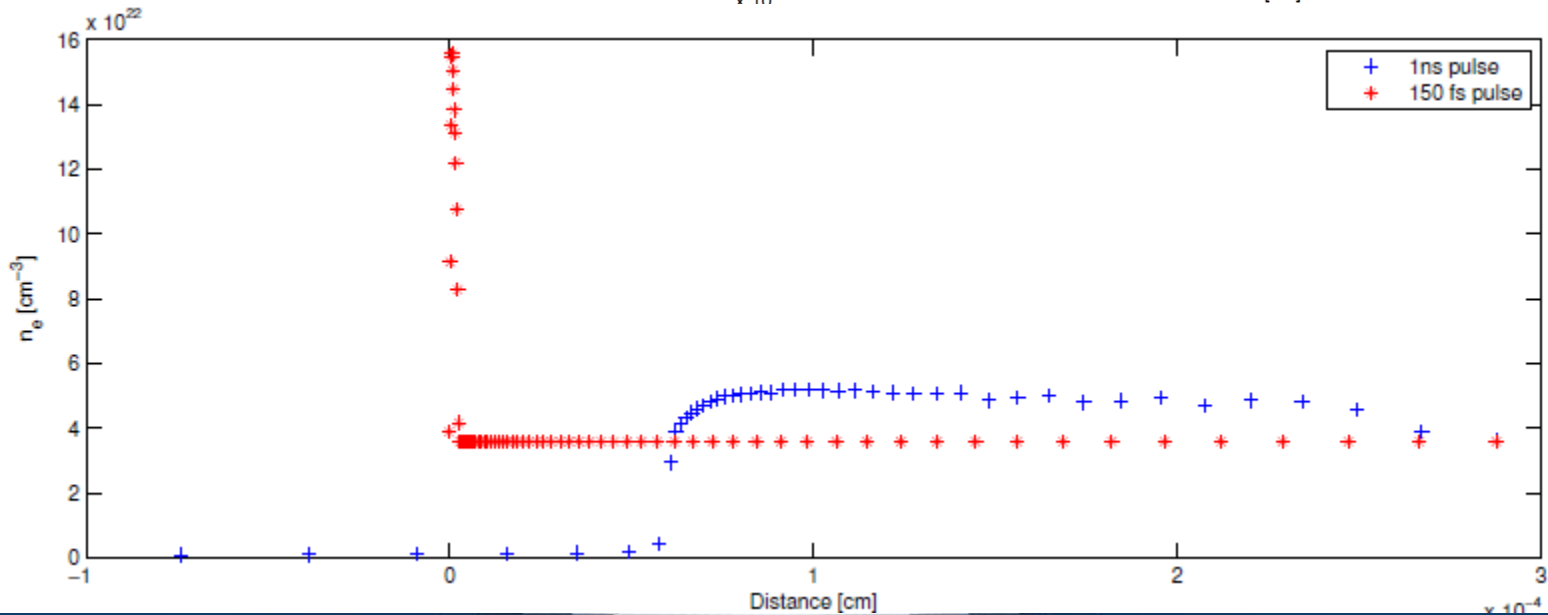
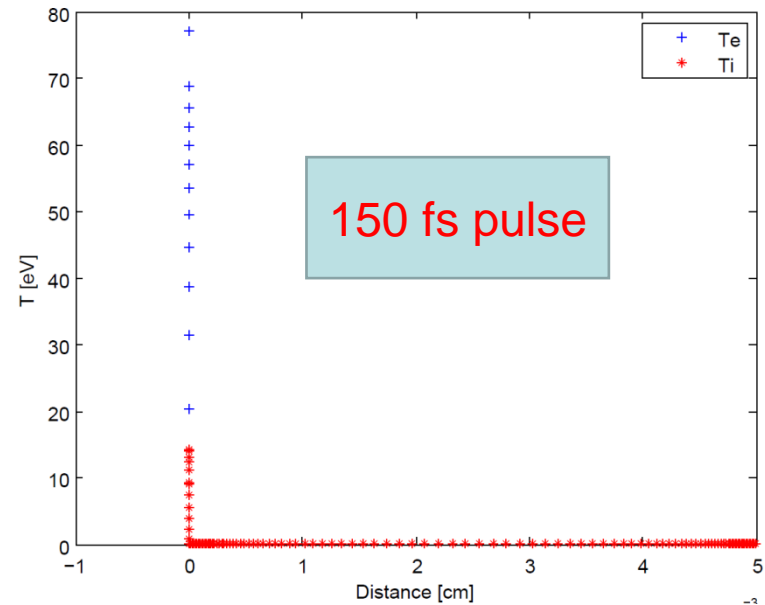
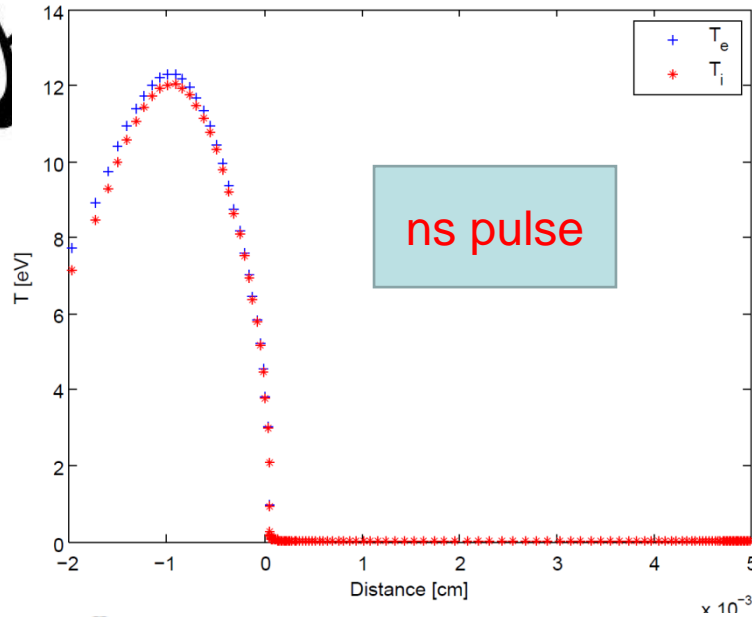
- **Density** 1.4 g/cm^3 .



- **Electron-ion collision and ion hydrodynamic expansion time are in sub ps.**



ns versus fs pulses II





Laser Droplet interaction movie

Entry #: V0016

Laser impact on a drop

Alexander L. Klein¹, Claas Willem Visser¹, Wilco Bouwhuis¹,
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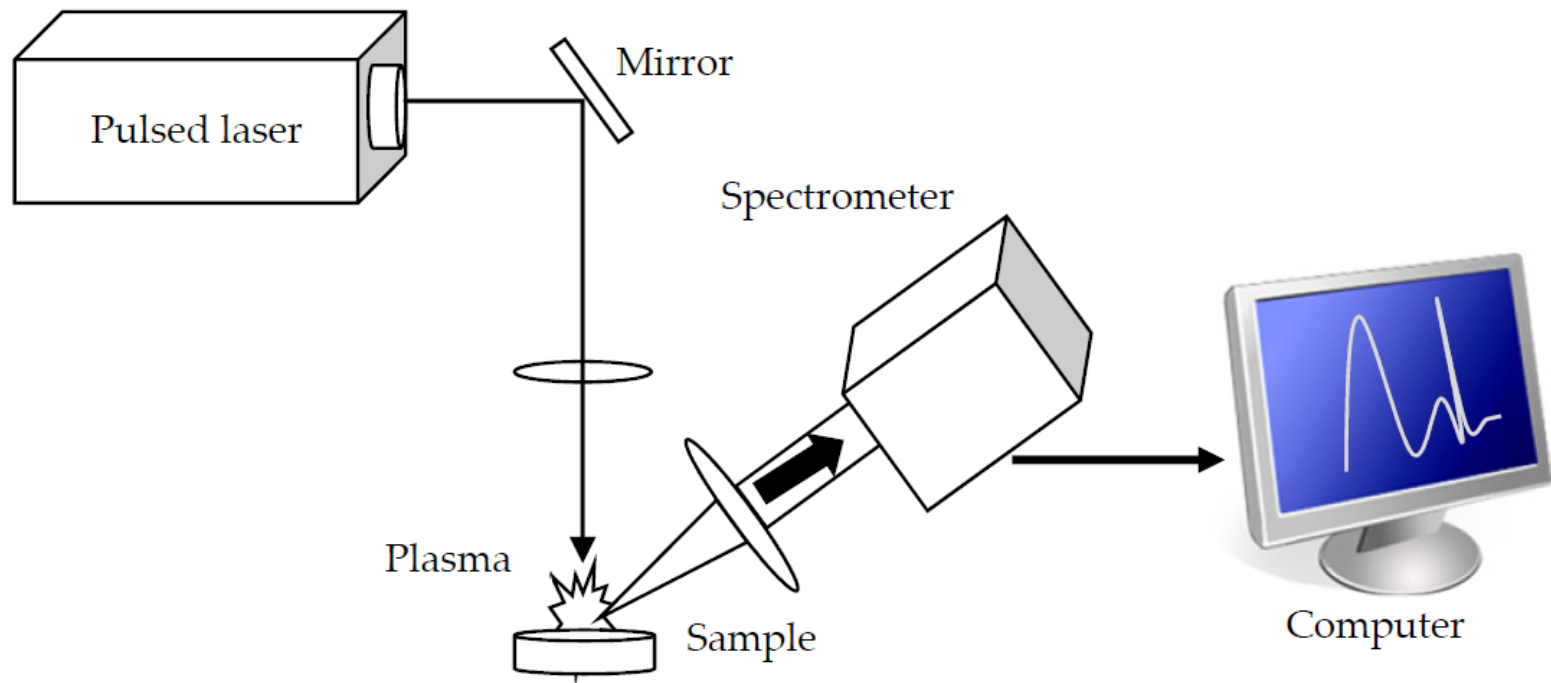




Applications



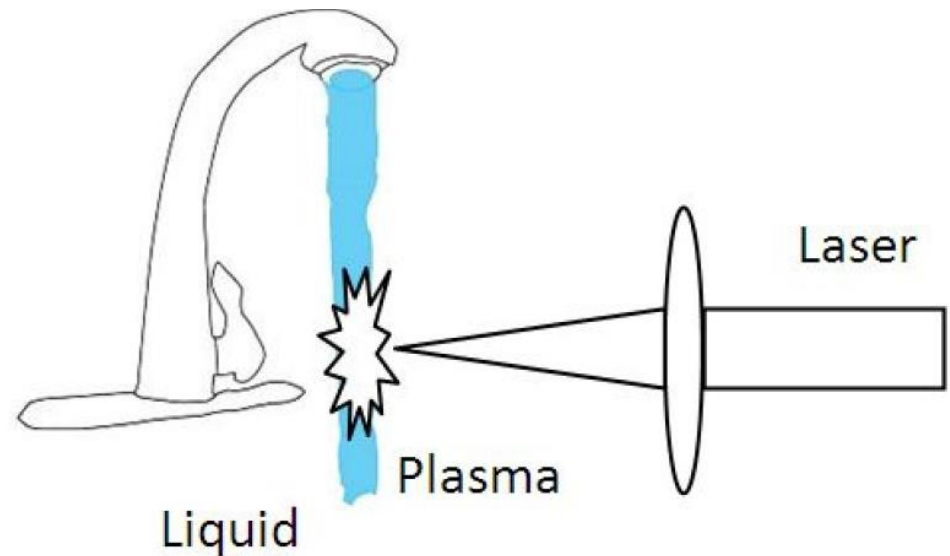
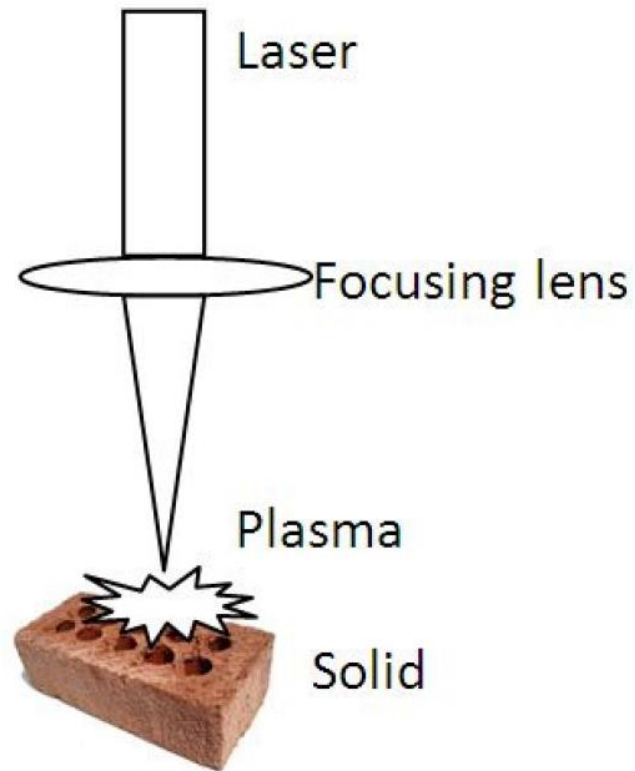
Laser induced breakdown spectroscopy



- **The conventional LIBS configuration**



LIBS for different Materials

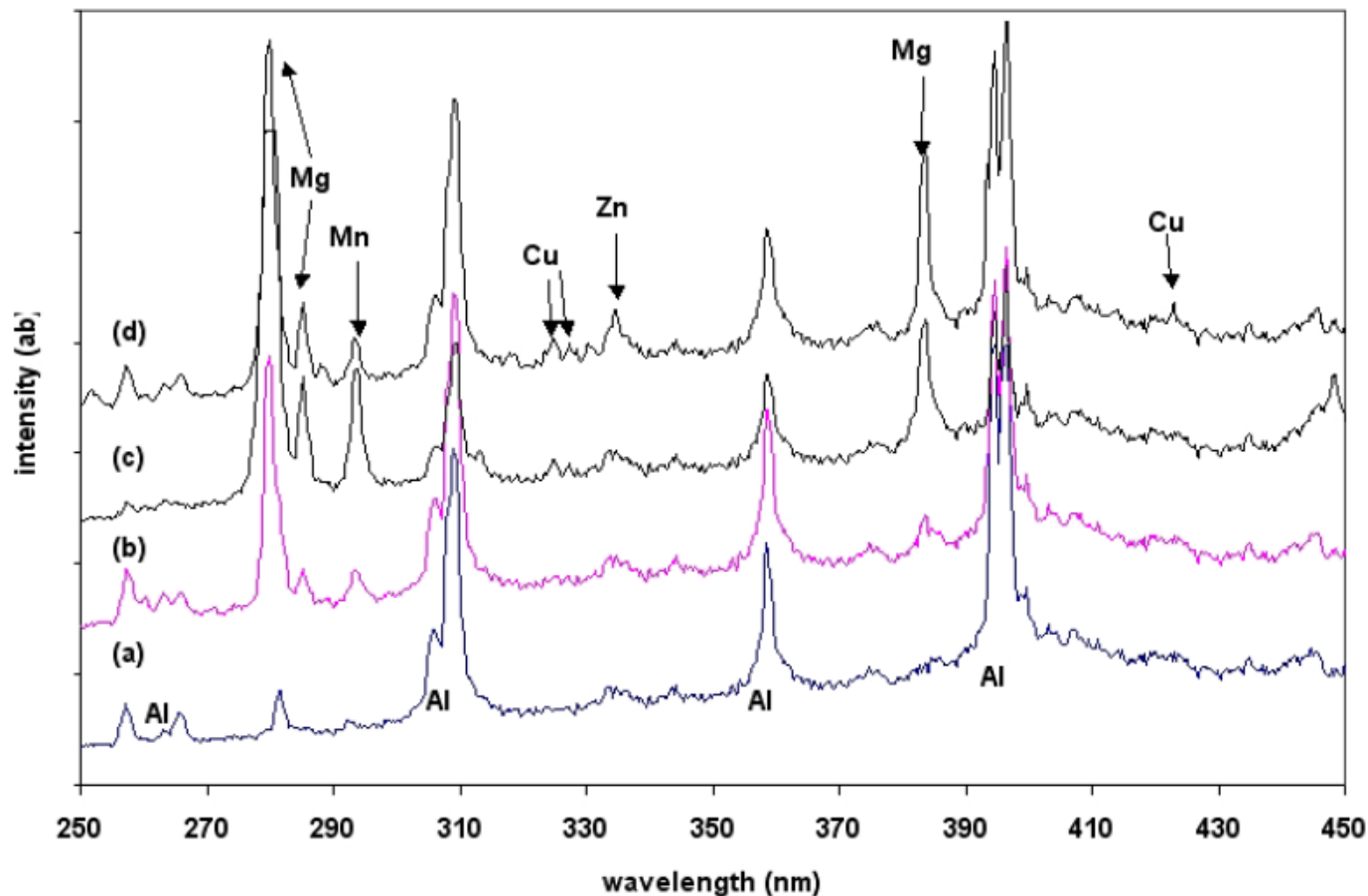


- Solid and liquid samples

InTech



Al alloys spectrum

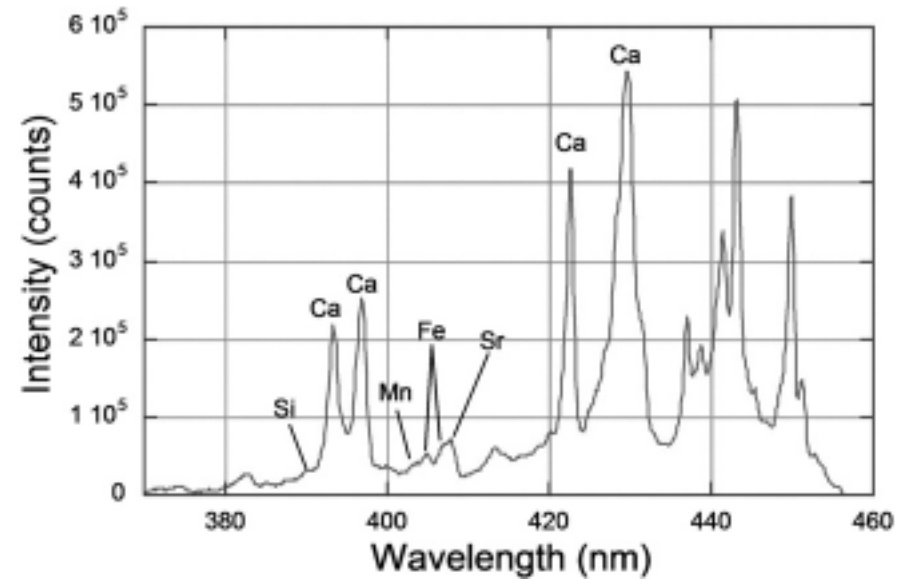
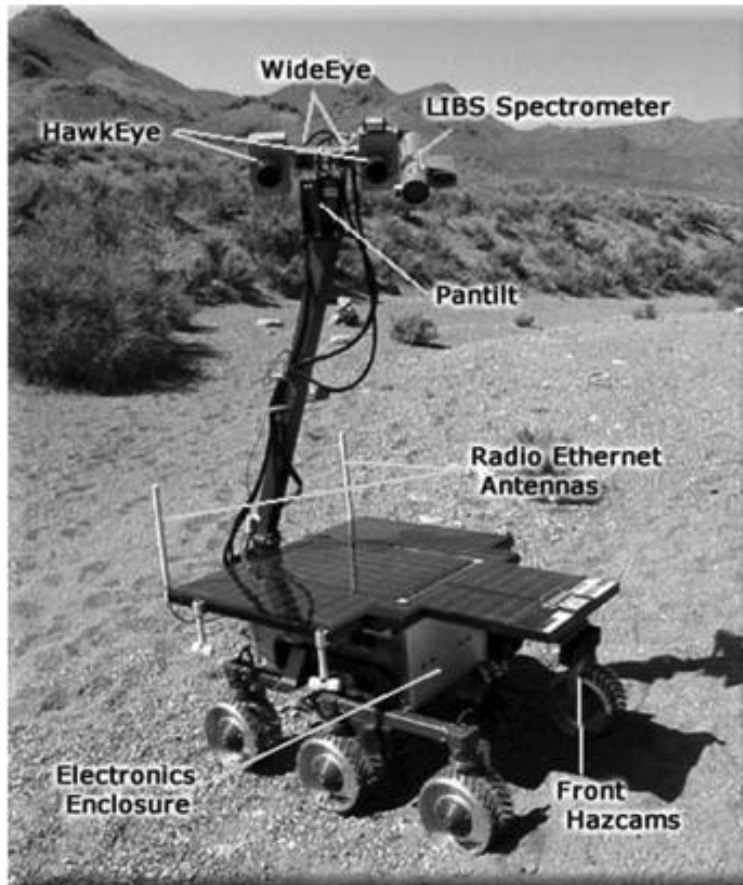


- LIBS for Al alloys (a) Pure Al, (b) 3003 alloy, (c) 2024-T3 alloy, and (d) 7057-T6 alloy.

InTech



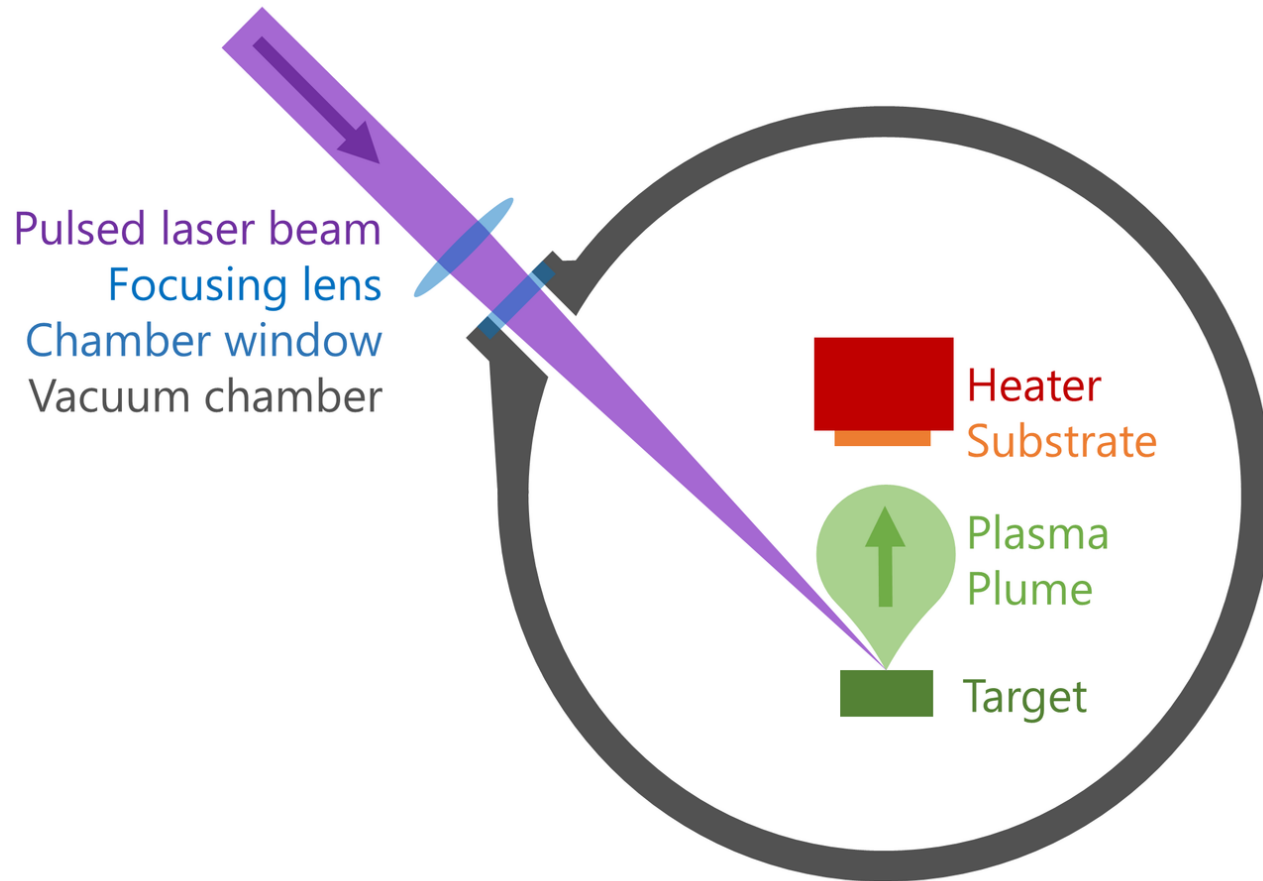
Study your samples in the field!!



- The k9 rover in the field (Nevada, USA).



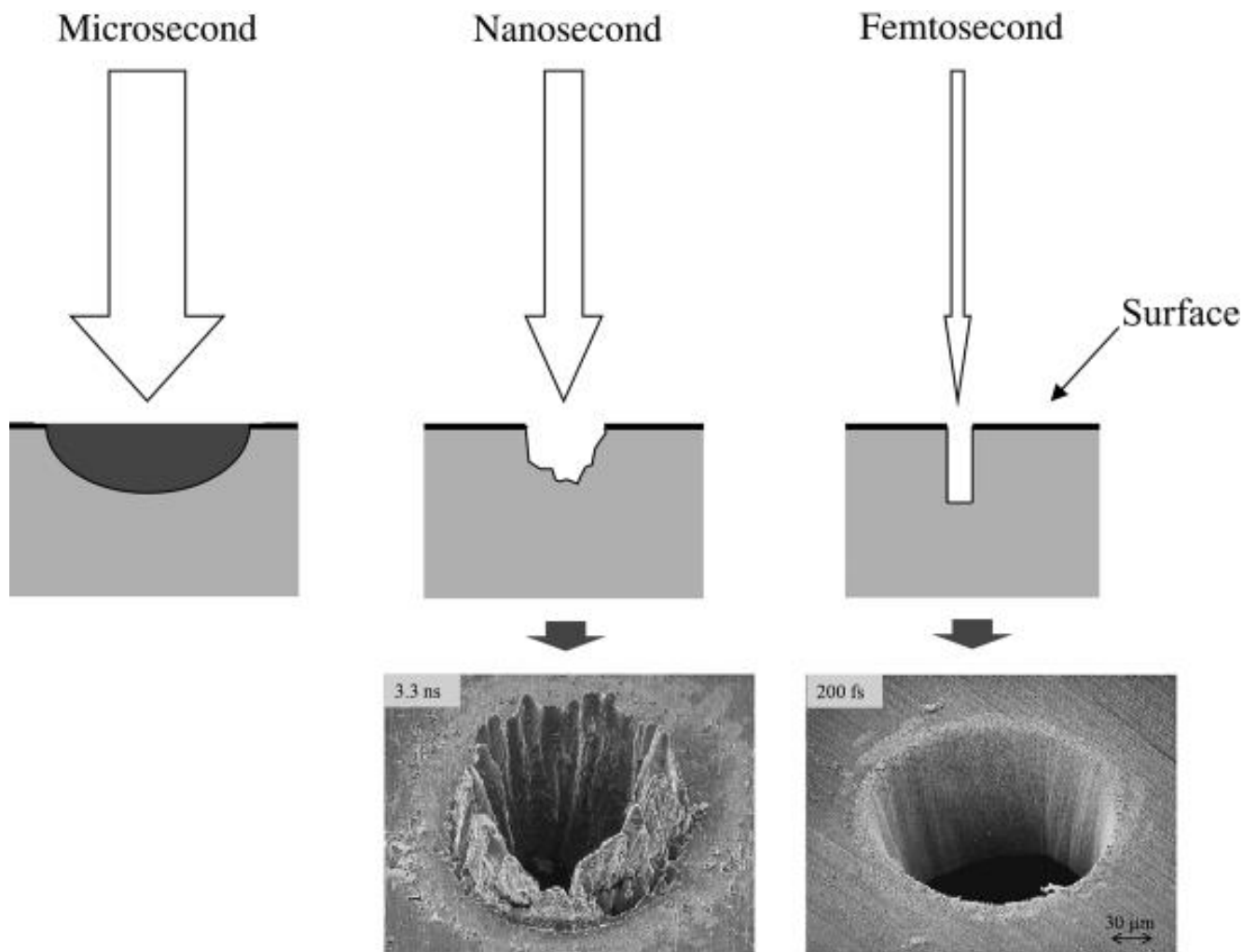
Thin film deposition



[WIKI](#)



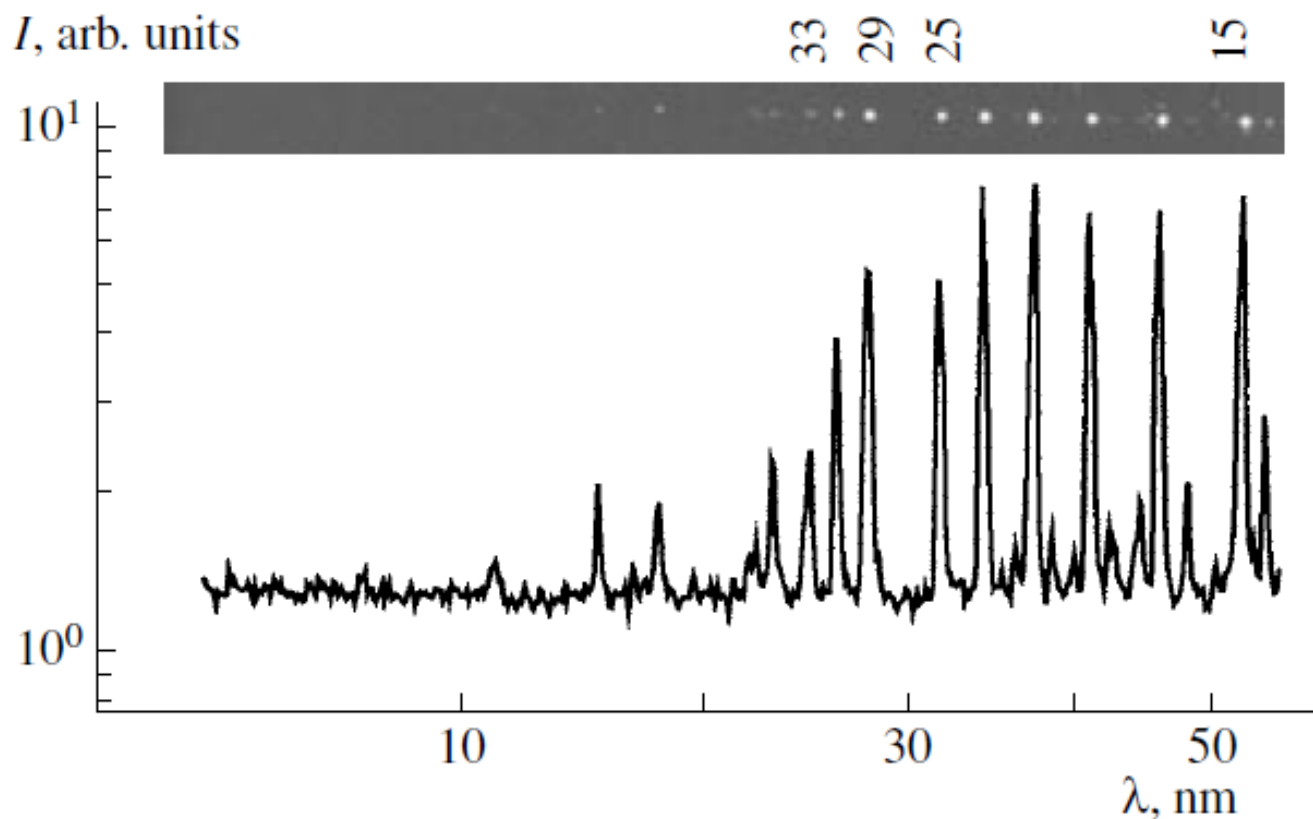
Plasma etching



Handbook of LIBS, Cremers and Radziemski



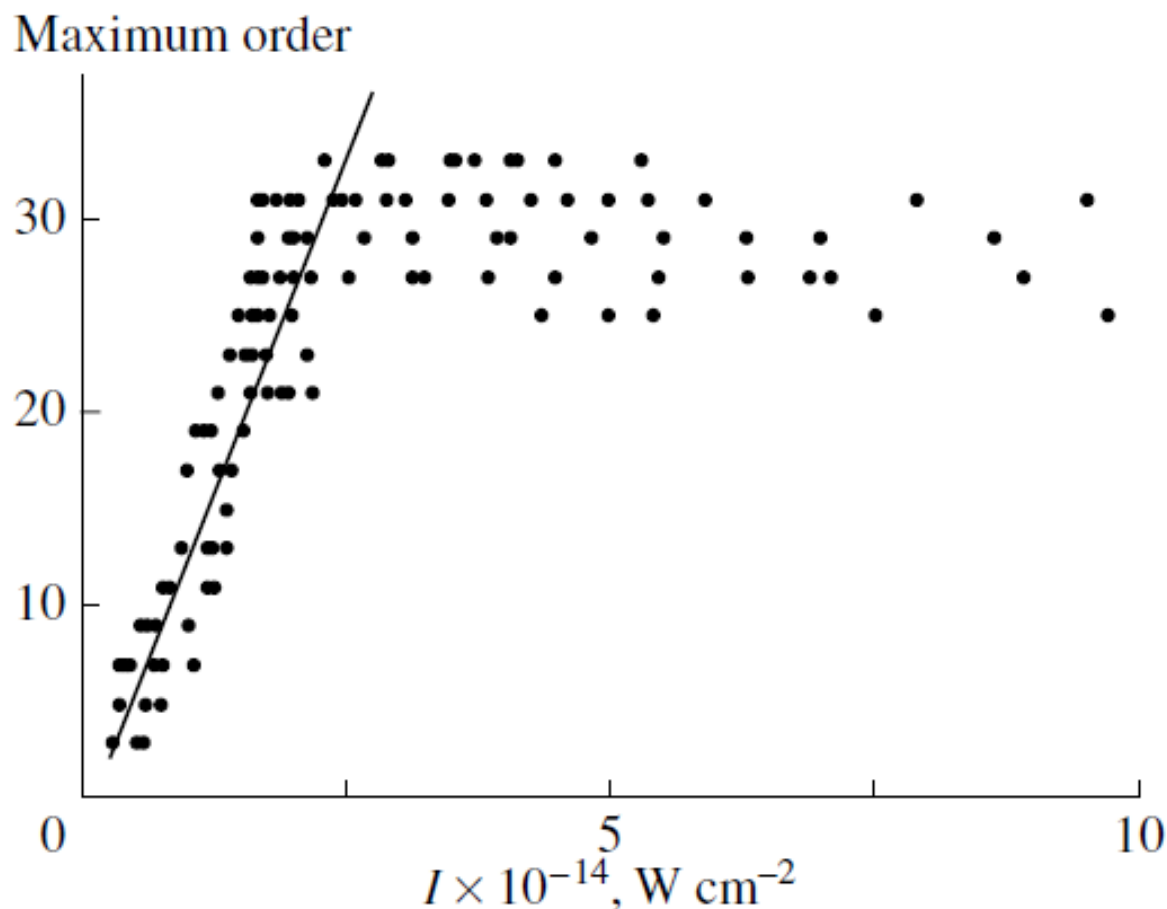
High harmonics generation I



Ganeev and Kuroda, optics and spectroscopy 2006



High harmonics generation I



Ganeev and Kuroda, optics and spectroscopy 2006



X-ray Production

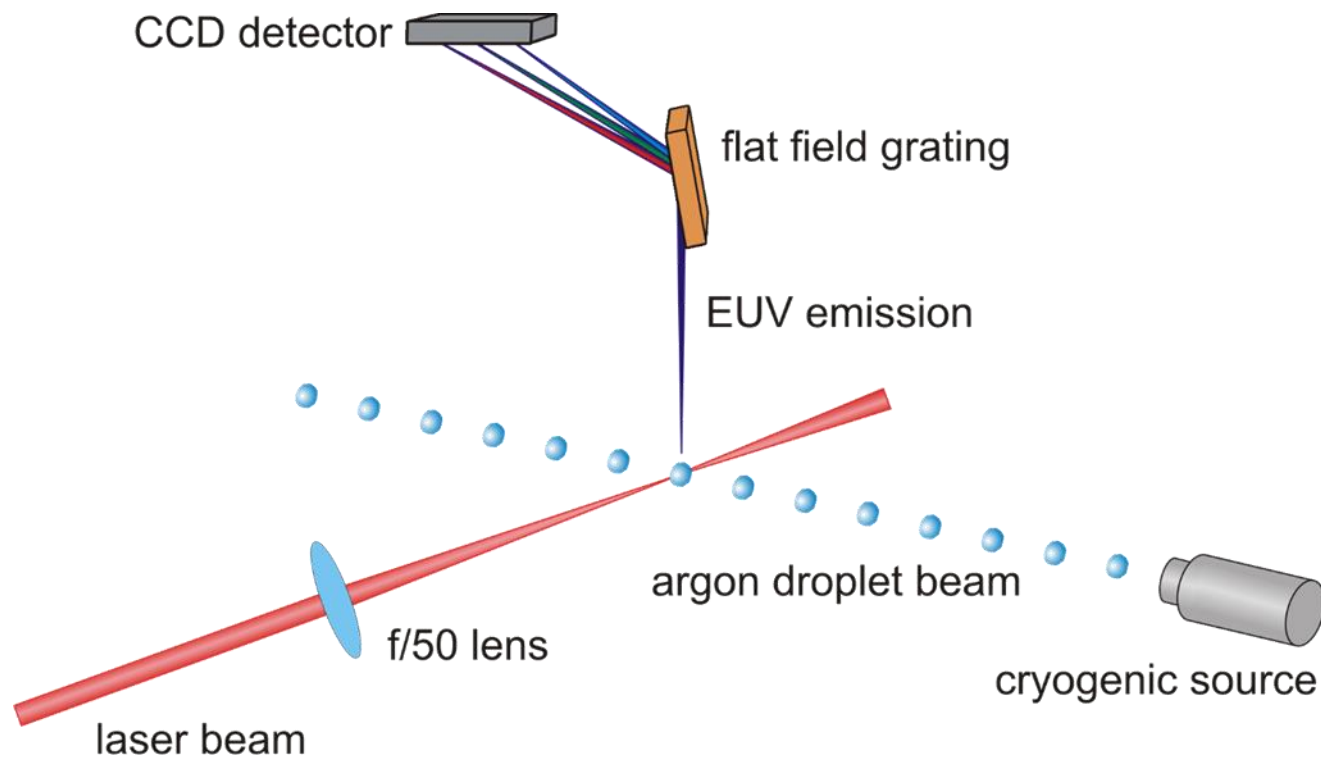
- Laser produced hot plasmas which can produce x-ray.
- Three mechanisms are exist:
 - Bremsstrahlung or (Free-Free emission)
 - Recombination radiation (Free-bound emission).
 - Line radiation (bound-bound emission).
- The laser to x-ray conversion efficiency E_x/E_L depends on the target atomic number, the laser intensity, the laser wavelength;

$$E_x/E_L = 6.3 \times 10^{-10} Z \lambda_{\mu m}^{-0.48} \left(\frac{I_0}{1 + I_0^2} \right)^{0.46}$$

H.C. Pant, plasma physics 1992



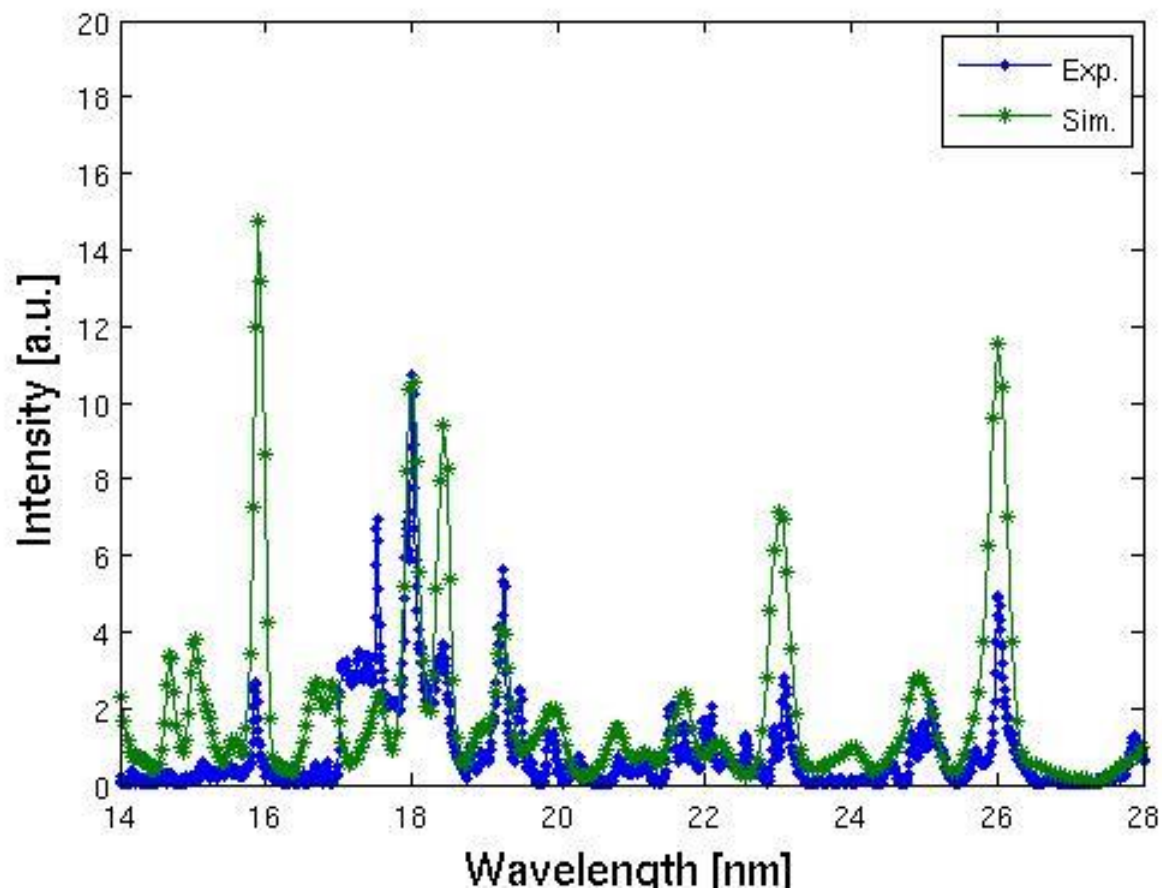
UV Source I



R. Irsig, M. Shihab et al 2018



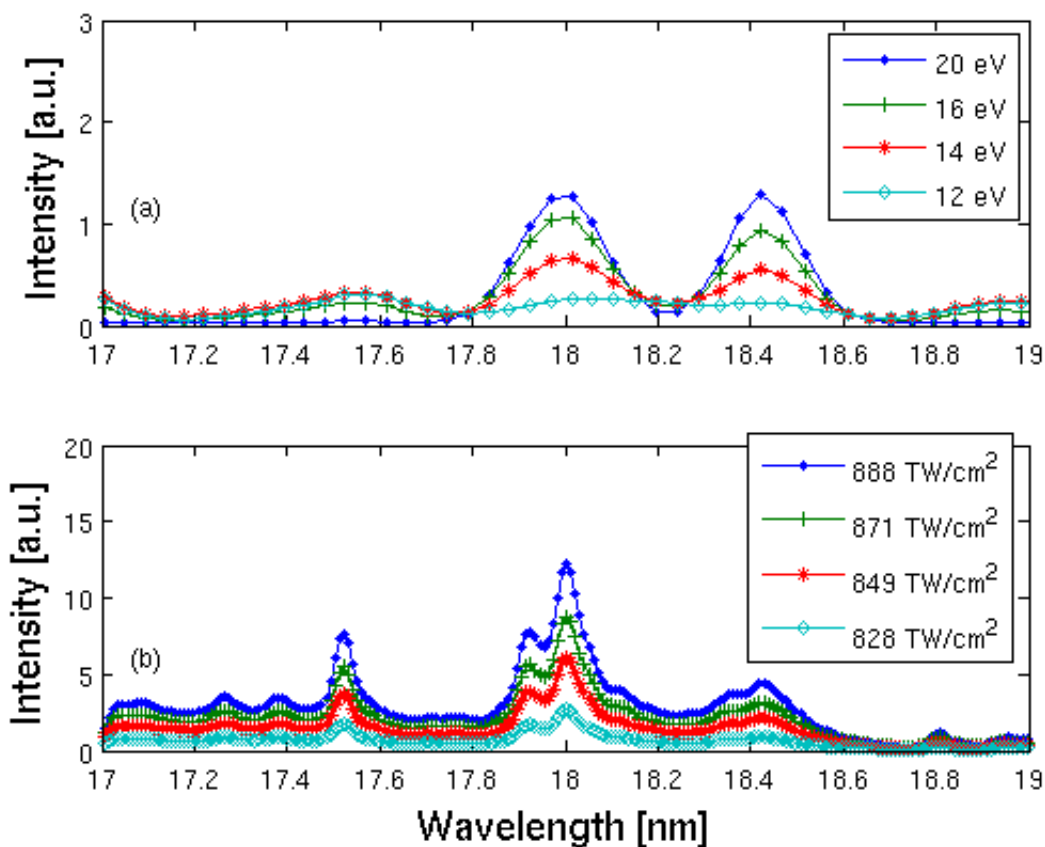
UV Source II



R. Irsig, M. Shihab et al 2018



UV Source III



R. Irsig, M. Shihab et al 2018

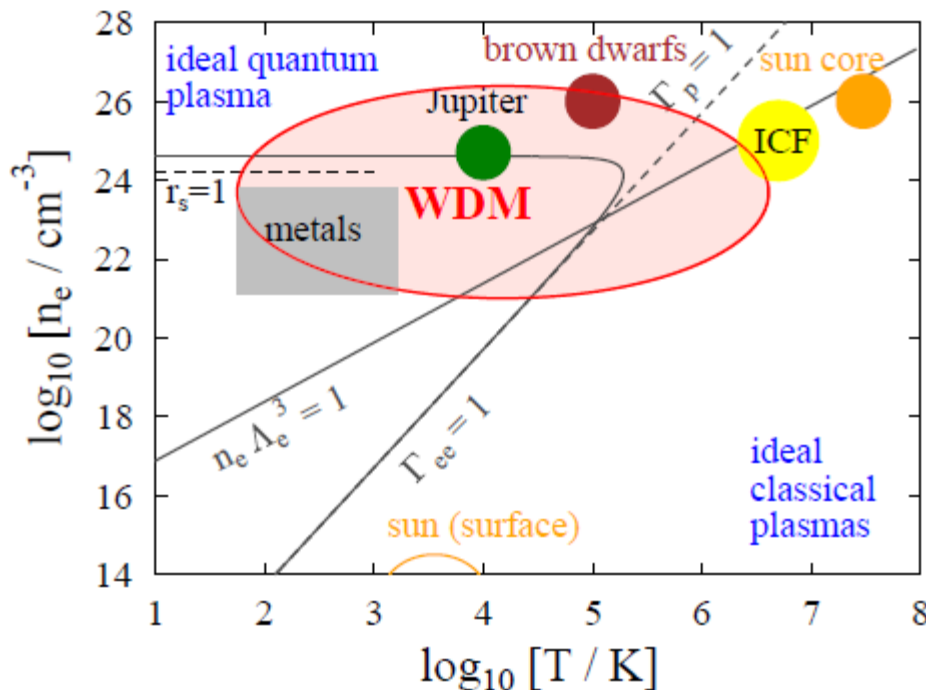


Metal cutting and welding





Warm Dense Matter



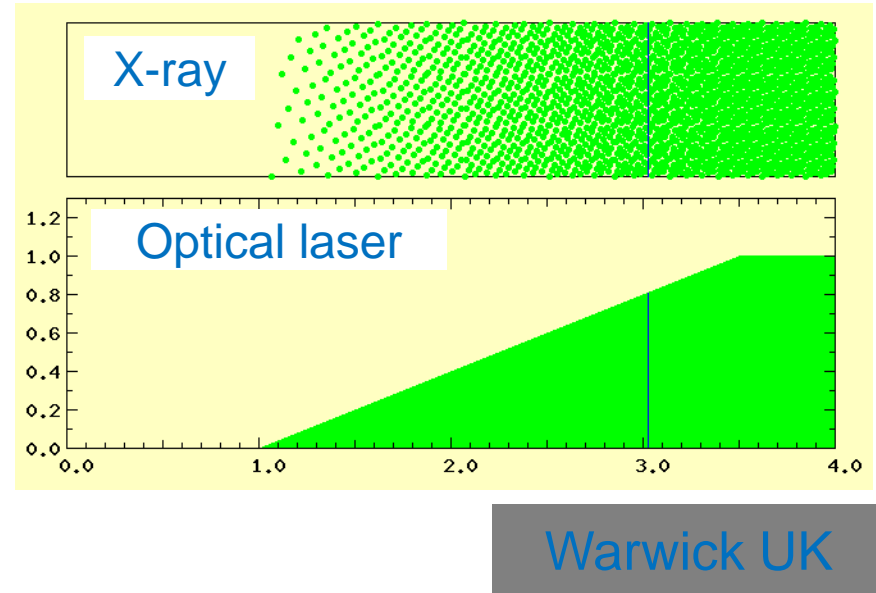
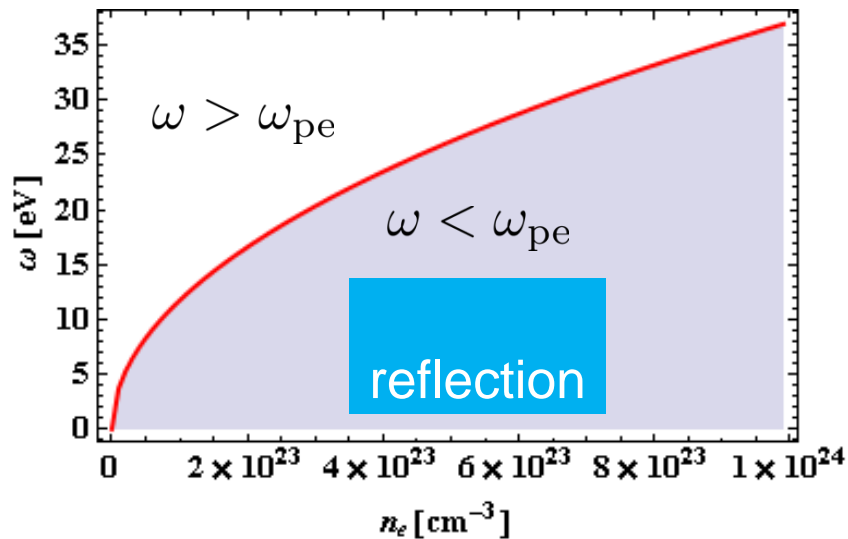
K. Wünsch

- WDM:
 - Temperature of few electronvolts
 - Solid state density and beyond
- ICF, shock experiments, giant planets, and brown dwarfs
- Theories of solid, condensed matter, or ideal plasma are not valid
- No single theoretical model describes the behavior of WDM
 - Partial ionization
 - Arbitrary degeneracy
 - Strong ionic correlations

Glenzer et al PRL 98 065002(2007)



Dispersion relation of EM in WDM

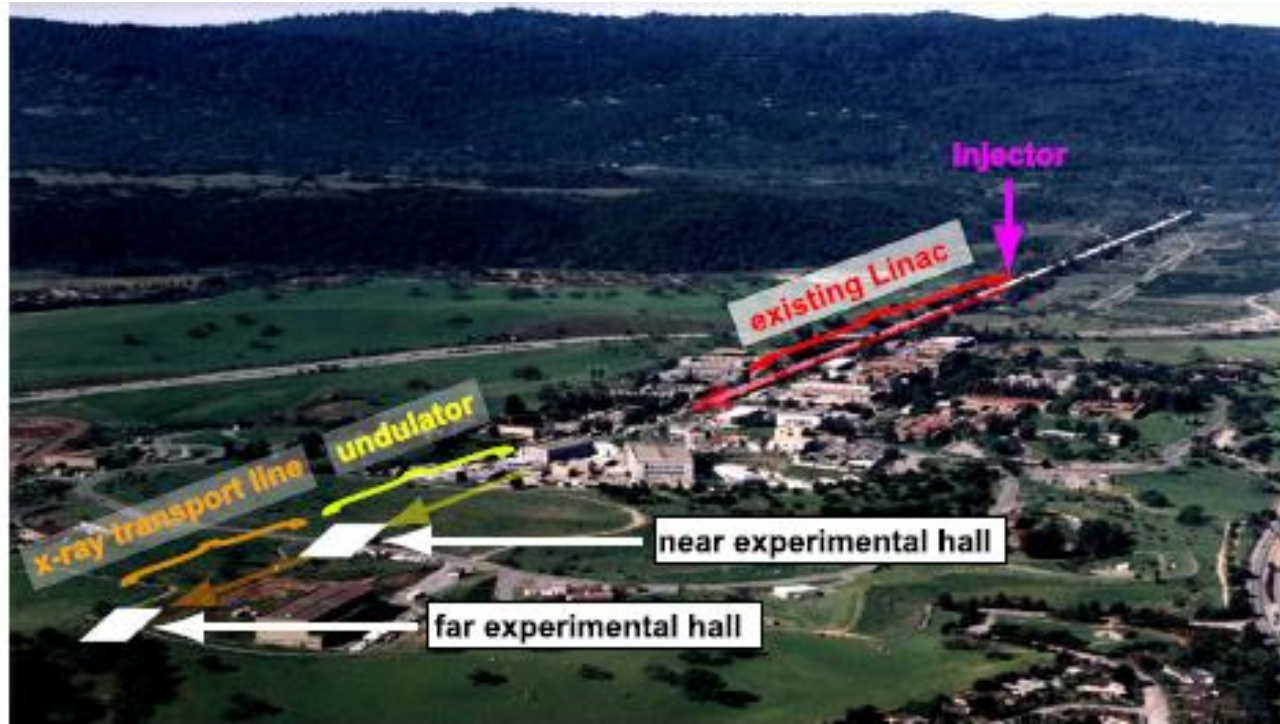


Warwick UK

- At the natural plasma oscillation: $\omega_{pe} = \omega \rightarrow k = 0$
- At the cut off, the wave is reflected: $\omega_{pe} > \omega \rightarrow k = i\kappa$
- WDM is transparent in x-ray regime:

$$n_e = 10^{24} \text{cm}^{-3} \rightarrow \lambda \leq 33 \text{nm}$$

Set-up of an XRTS experiment

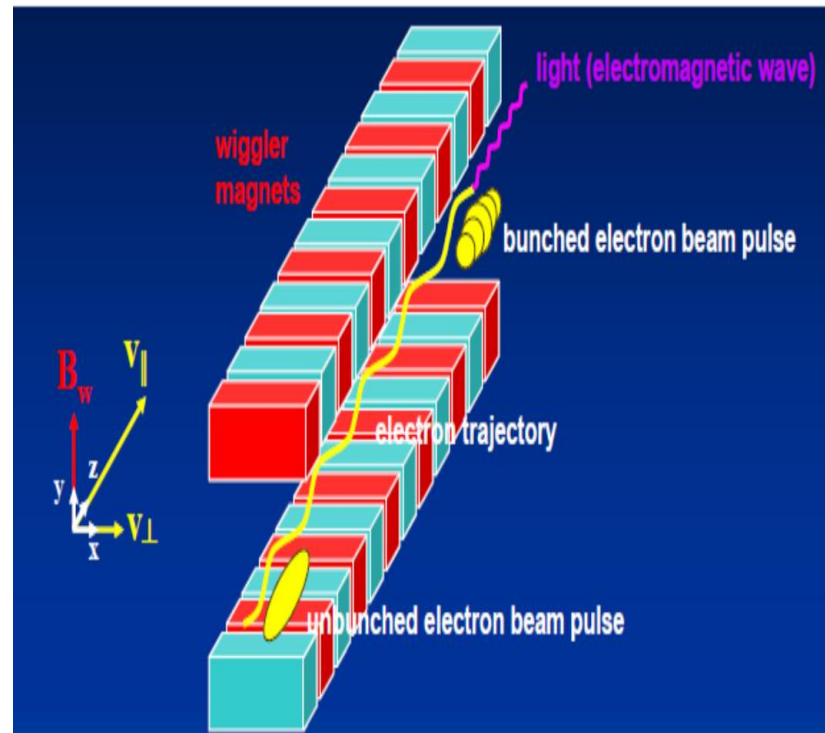


S.H. Glenzer et al., (2016): Stanford University



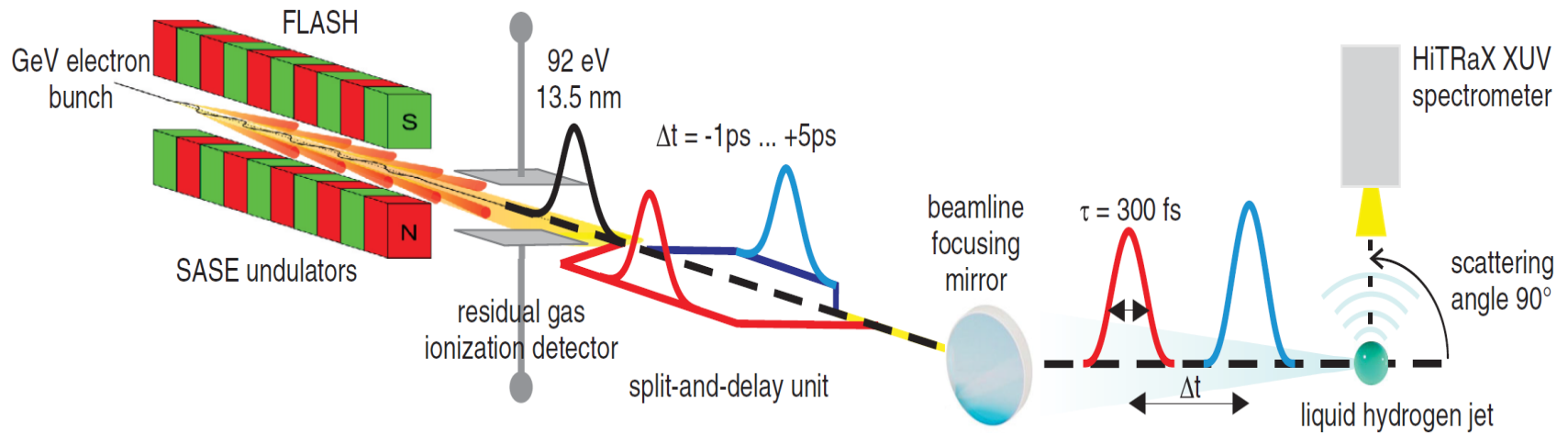
Free electron laser

- ❖ The free electron laser (FEL) is a device that transforms the kinetic energy of a relativistic electron beam into electromagnetic (EM) radiation.
- ❖ Electrons in an FEL are not bound to atoms or molecules.
- ❖ The “free” electrons traverse a series of alternating magnets, called a “wiggler,” and radiate light at wavelengths depending on electrons’ energy, wiggler period and magnetic field.





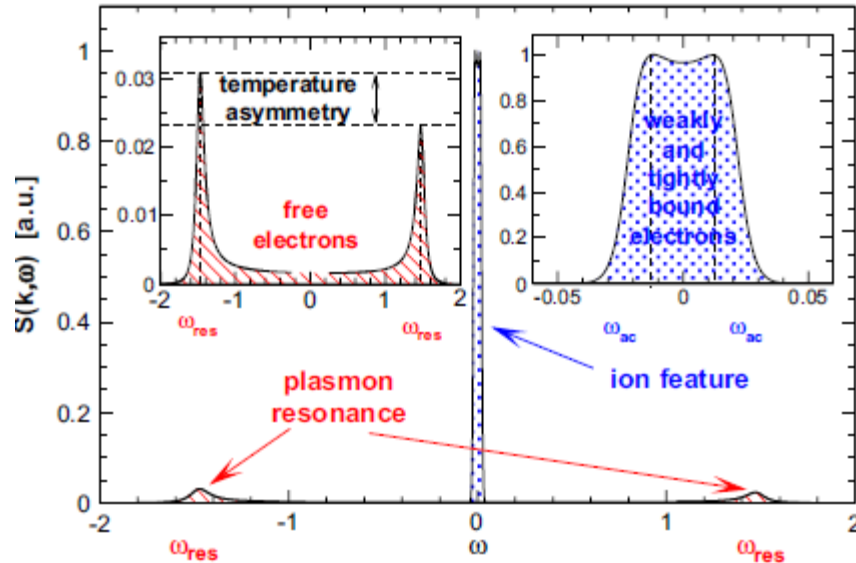
Time Delay experiment



U. Zastrau 2014: FLASH(Hamburg)



XRTS features



A. Höll et al., HEDP 3, 120(2007)

- Thomson scattering has two distinct features:

- Inelastic scattering (frequency shifted) from free electrons and bound free transitions
- Unshifted Rayleigh peak (elastic) due to electrons co-moving with the ions

- The electrons in partially ionized system can be split into bound and free electrons

$$\rho_e = \rho_b + \rho_f$$

- Intermediate scattering function

$$N_e F_{ee}^{tot} = \langle \rho_b(\vec{k}, t) \rho_b(-\vec{k}, t) \rangle + 2 \langle \rho_f(\vec{k}, t) \rho_b(-\vec{k}, t) \rangle + \langle \rho_f(\vec{k}, t) \rho_f(-\vec{k}, t) \rangle$$



Born-Mermin approximation

- Fluctuation-dissipation theorem :

$$S_{ee}^0(k, \omega) = -\frac{\epsilon_0 \hbar k^2}{\pi e^2 n_e} \frac{\text{Im} \epsilon^{-1}(k, \omega)}{1 - \exp(-\hbar \omega / k_B T_e)}$$

- RPA given by Lindhard:

$$\epsilon^{\text{RPA}}(\vec{k}, \omega) = 1 - \frac{1}{\epsilon_0 \Omega_0 k^2} \sum_p e^2 \frac{f_{p+k/2}^e - f_{p-k/2}^e}{\Delta E_{p,k}^e - \hbar(i\omega + i\eta)}$$

- Mermin ansatz :

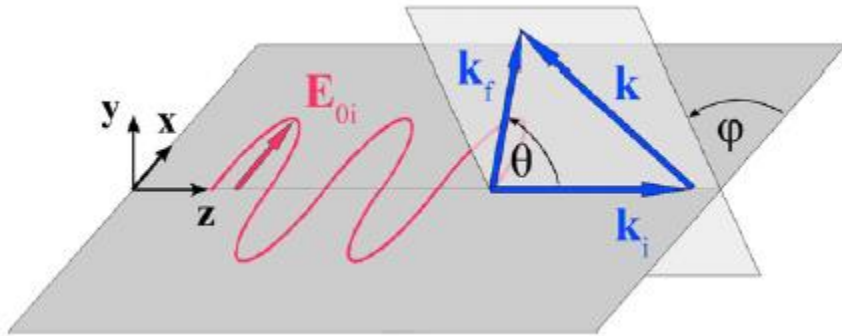
$$\epsilon_M(k, \omega) = 1 + \frac{\left(1 + \frac{i\nu(\omega)}{\omega}\right) [\epsilon^{\text{RPA}}(k, \omega + i\nu(\omega)) - 1]}{1 + i \frac{\nu(\omega)}{\omega} \frac{\epsilon^{\text{RPA}}(k, \omega + i\nu(\omega)) - 1}{\epsilon^{\text{RPA}}(k, 0) - 1}}$$

- $\nu(\omega)$ is the dynamic collision frequency via Born approximation.

Glenzer and Redmer, RMP 81, 1625(2009)



Back and forward scattering



- The momentum transfer depends on the scattering angle

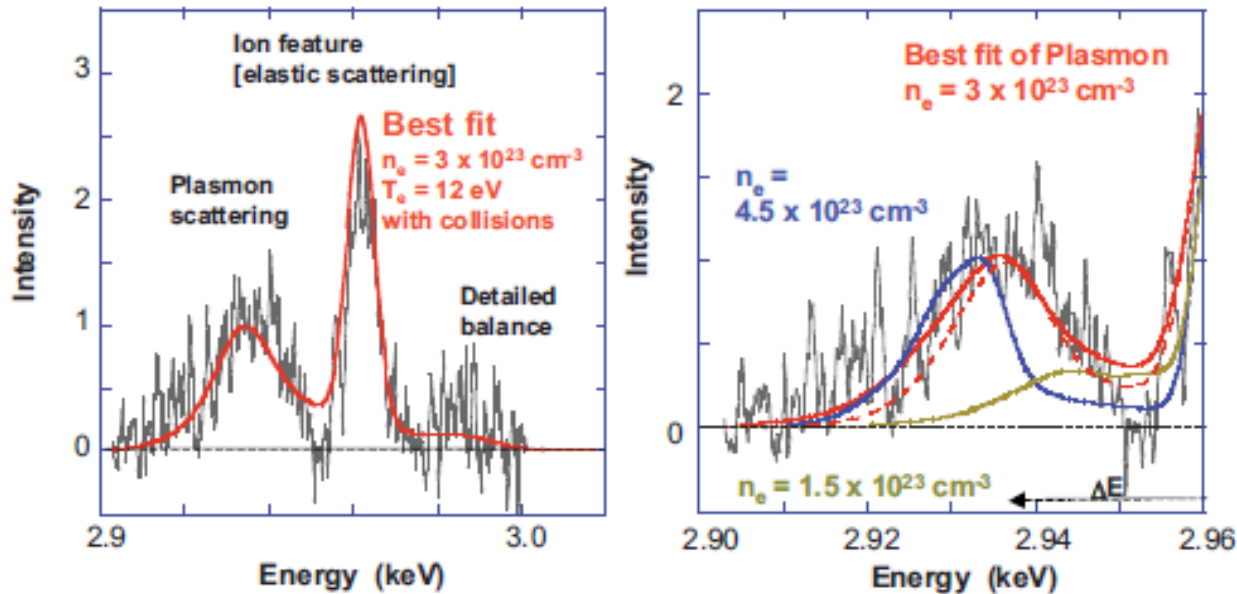
$$k = |k_f - k_i| = \frac{4\pi}{\lambda_i} \sin(\theta/2)$$

- Dimensionless scattering parameter $\alpha = \frac{1}{k\lambda_{sc}} = \frac{l}{2\pi\lambda_{sc}}$
 - l is the electron density fluctuation
 - λ_{sc} is the screening length
- Collective scattering: ($\alpha > 1$)
 - the scattering reflects the electron density fluctuations
 - Plasmon features
- Non-collective scattering: ($\alpha < 1$)
 - the scattering reflects the velocity distribution of electrons
 - Compton features

Glenzer and Redmer, RMP 81, 1625(2009)



Experimental results and

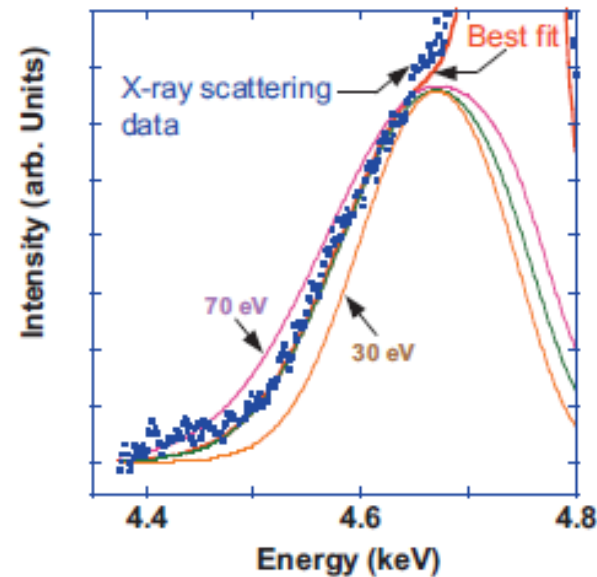
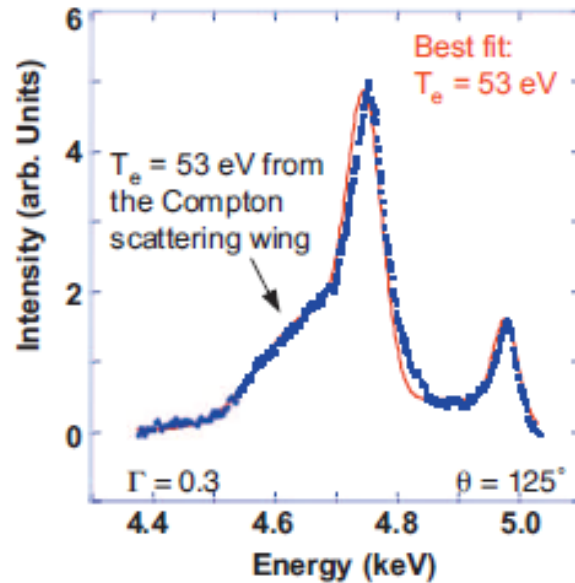


- **Forward scattering:** collective behavior
- Dispersion relation determines the electron density
- Detailed balance gives the electron temperature

Glenzer et al., PRL 98, 065002(2007)



Experimental results and synthetic spectra II



- **Back scattering:**
 - Compton scattering
 - Non-collective behavior
 - Line width \propto Fermi energy

Glenzer et al., PRL 90, 175002(2003)



Thanks!



Collisional Radiative Model

Assumptions:

- Homogeneous plasmas
- The expansion velocity is constant

$$v_{\text{exp}} = \text{const} \quad N(r, t) = N(r', t')(t'/t)^3$$

- The relative density of charge state is given by

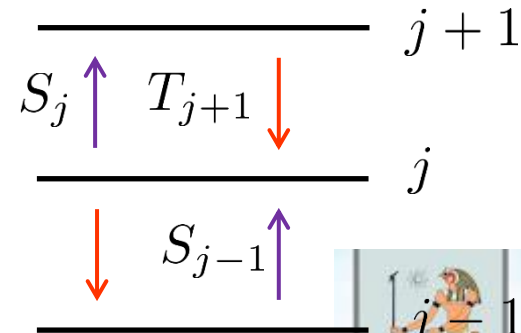
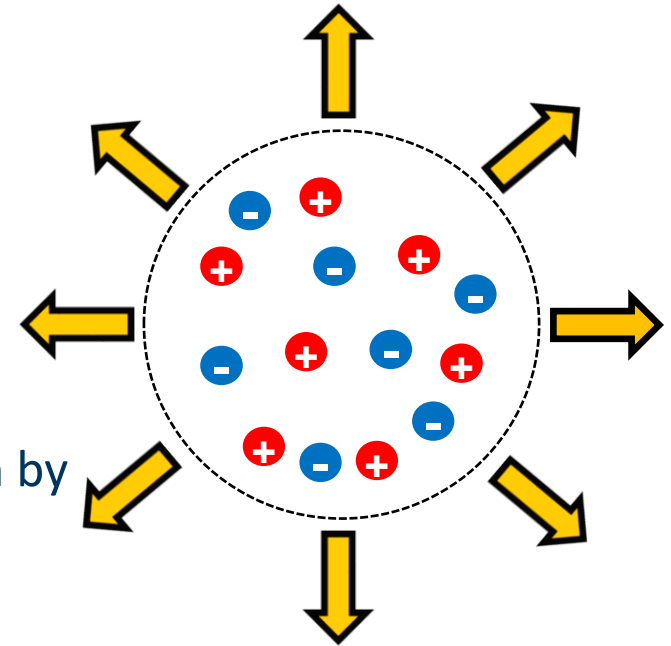
$$n_j = \frac{N_j}{N_T}$$

Governing equations:

$$\frac{\partial n_j}{\partial t} = N_e S_{j-1} n_{j-1} + N_e T_{j+1} n_{j+1} - N_e (S_j + T_j) n_j$$

$$\frac{\partial T_e}{\partial t} = \frac{-2T_e}{t} + \frac{2}{3N_e} (P_{\text{recom}} - P_{\text{brem}} - P_{\text{ioniz}}) - P_{\text{ei}}$$

$$\frac{\partial T_i}{\partial t} = \frac{-2T_i}{t} + P_{\text{ei}}$$





Collisional Radiative Model

- **Ionization by collisions**

$$S(Z) = 2.43 \times 10^{-6} \xi_Z T_e^{3/2} \exp(-u) / u^{7/4} \text{ cm}^3 \text{ s}^{-1}$$

- **Recombination:** $T_j = \alpha_j + \beta_j + D_j$

Radiative + Three body + Dielectronic

$$\alpha(Z + 1) = 5.2 \times 10^{-14} (Z + 1) u^{1/2} (0.429 + 0.5 \ln u + 0.469 u^{-1/2}) \text{ cm}^{-3} \text{ s}^{-1}$$

$$D(Z + 1) = \frac{1}{T_e^{3/2}} \sum_{j=2}^{Z-1} \sum_{m=1}^4 C_{mj} \exp(-E_{mj}/T_e)$$

$$\beta(Z + 1) = 8.75 \times 10^{-27} N_e (Z + 1)^3 / T_e^{9/2} \text{ cm}^{-3} \text{ s}^{-1}$$



Collisional Radiative Model

- **Bremsstrahlung-radiation**

$$P_{\text{brem}} = 9.6 \times 10^{-14} \bar{Z} N_e^2 T_e^{1/2} \text{ eV s}^{-1} \text{ cm}^{-3}$$

- **Ionization energy loss**

$$P_{\text{ioniz}} = \sum_{j=1}^{nZ} nZ - 1 N_e N_j S_j \chi_Z \text{ eV s}^{-1} \text{ cm}^{-3}$$

- **Energy equilibrium**

$$P_{\text{ei}} = \frac{T_e - T_i}{\tau_{\text{eq}}} \text{ eV s}^{-1}$$

- **Three-body recombination**

$$P_{\text{recom}} = \sum_{j=2}^{nZ} N_e N_j \beta_j \chi_Z \text{ eV s}^{-1} \text{ cm}^{-3}$$

- **Continuum Lowering**



Non-equilibrium Nickel plasmas

- Nickel is a transition element.
- Nickel plays an essential role for the preparation of special stainless-steels and alloys to the production of antimagnetic screens and chemical reactive films.

D.B. Chrisey, G.K. Hubler, Pulsed Laser Deposition of thin films, Wiley, 1994

S.S. Harilal et al J. Appl. Phys. 114, 203302 (2013)

L. Torrisi et al Plasma Phys. Rep. 34, 547 (2008)

S. Amoruso et al, Appl. Phys. A. 89, 1017 (2007)

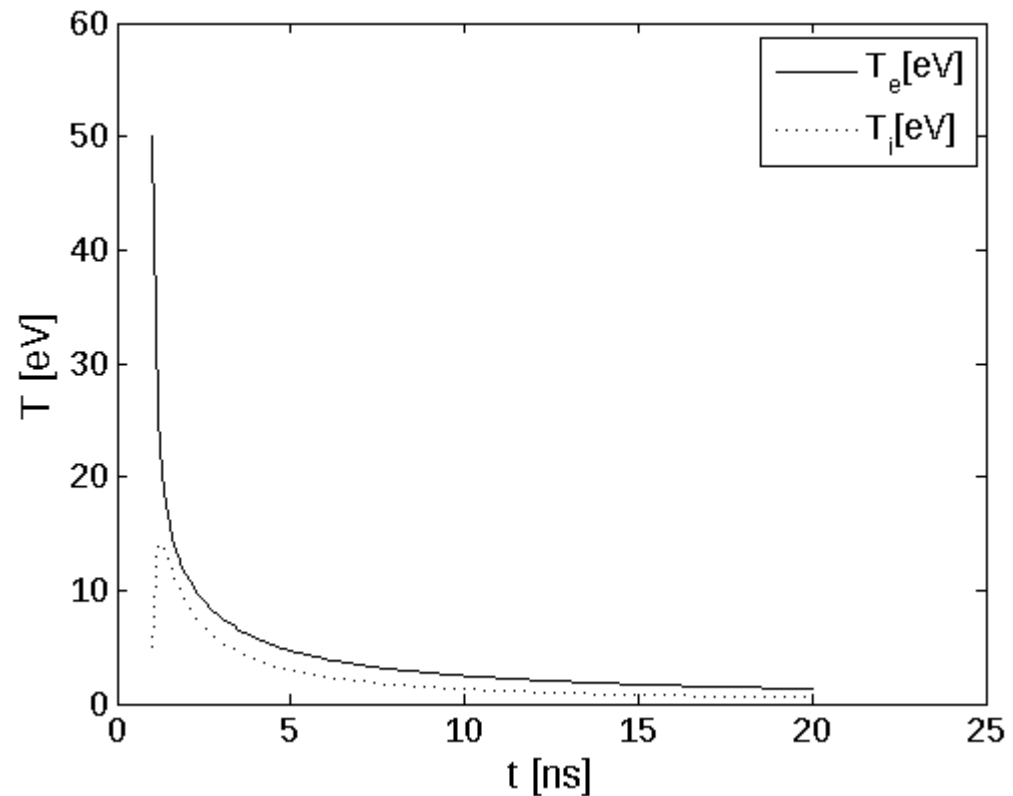


Electron-ion temperature

$$N = 10^{18} \text{ cm}^{-3}$$

$$T_e = 50 \text{ eV}$$

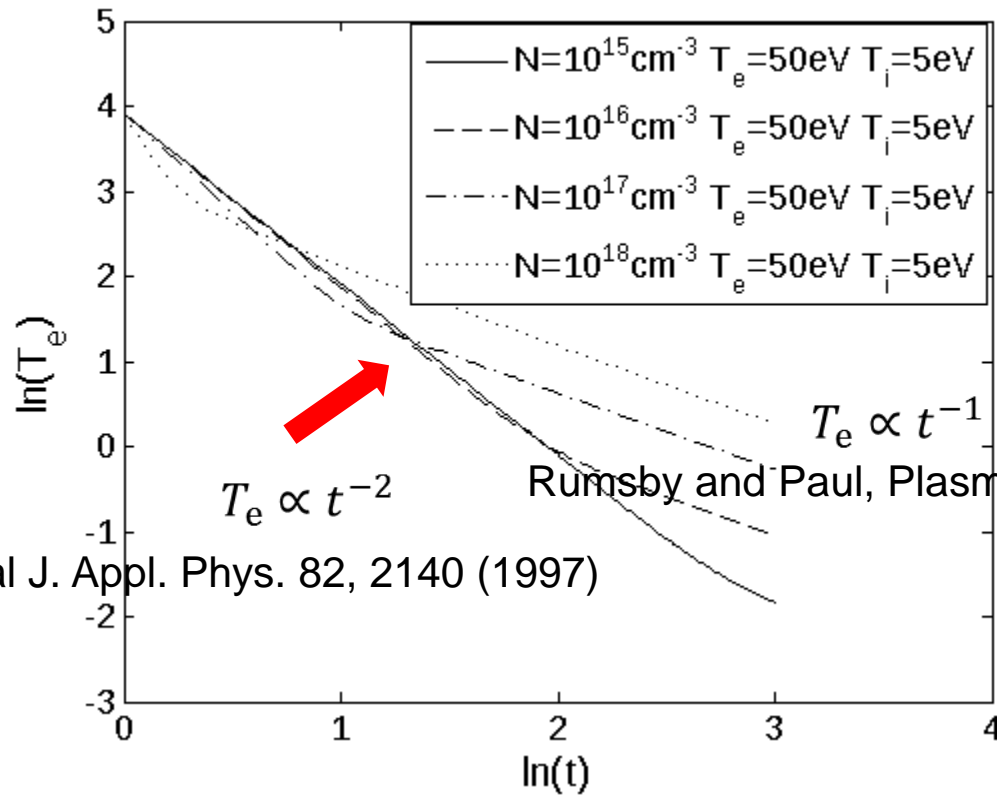
$$T_i = 5 \text{ eV}$$



M. Shihab, G.H. Abou-koura, N.M. El-Siragy, Appl. Phys. B (2016) 122-146



Electron temp.



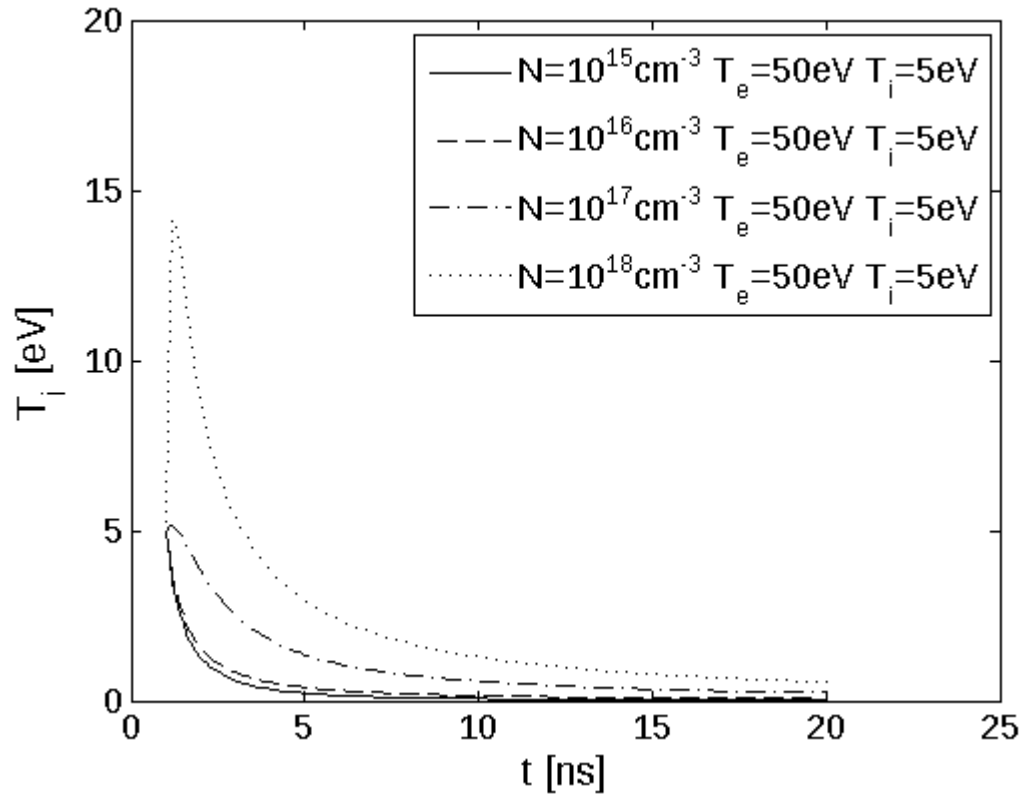
Rumsby and Paul, Plasma Phys. 16, 247 (1970)

S.S. Harilal et al J. Appl. Phys. 82, 2140 (1997)

$$\frac{\partial T_e}{\partial t} = \frac{-2T_e}{t} + \frac{2}{3N_e}(P_{\text{recom}} - P_{\text{brem}} - P_{\text{ioniz}}) - P_{\text{ei}}$$

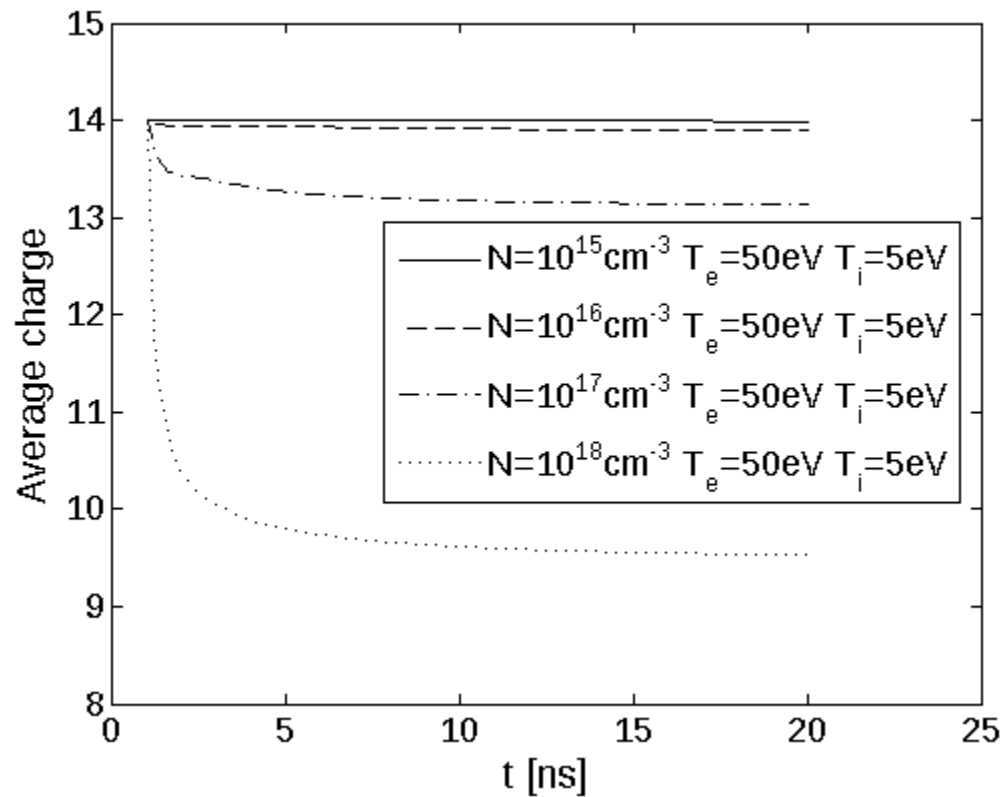


Ion temp.



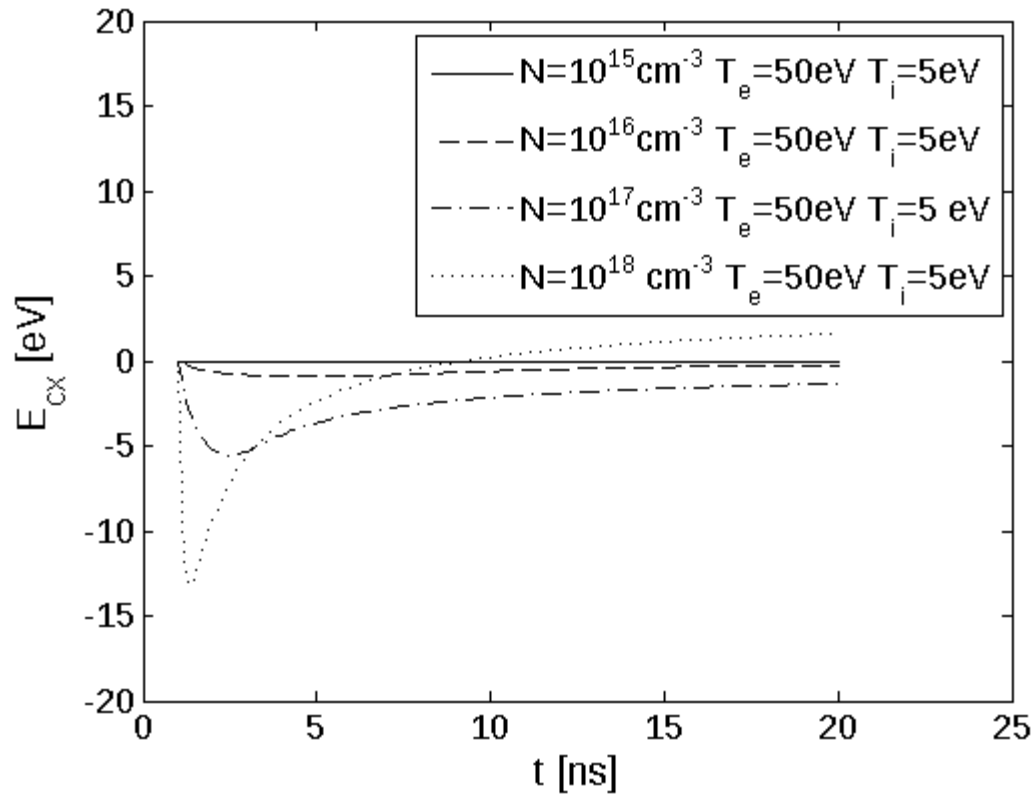


Average charge





Energy exchange



$$\frac{\partial T_e}{\partial t} = \frac{-2T_e}{t} + \frac{2}{3N_e} (P_{\text{recom}} - P_{\text{brem}} - P_{\text{ioniz}}) - P_{\text{ei}}$$

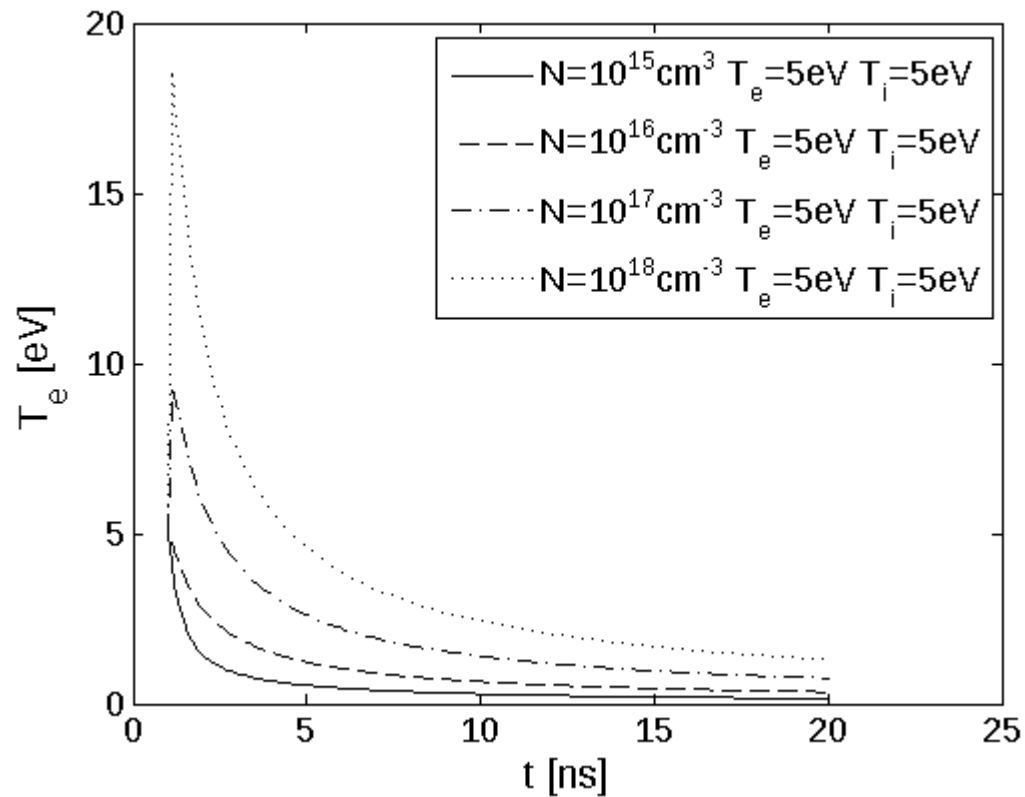
$$\frac{\partial T_i}{\partial t} = \frac{-2T_i}{t} + P_{\text{ei}} \quad \frac{\partial E_{\text{CX}}}{\partial t} = \frac{2}{3N_e} P_{\text{recom}} - P_{\text{ei}}$$



Equilibrium Ni plasmas

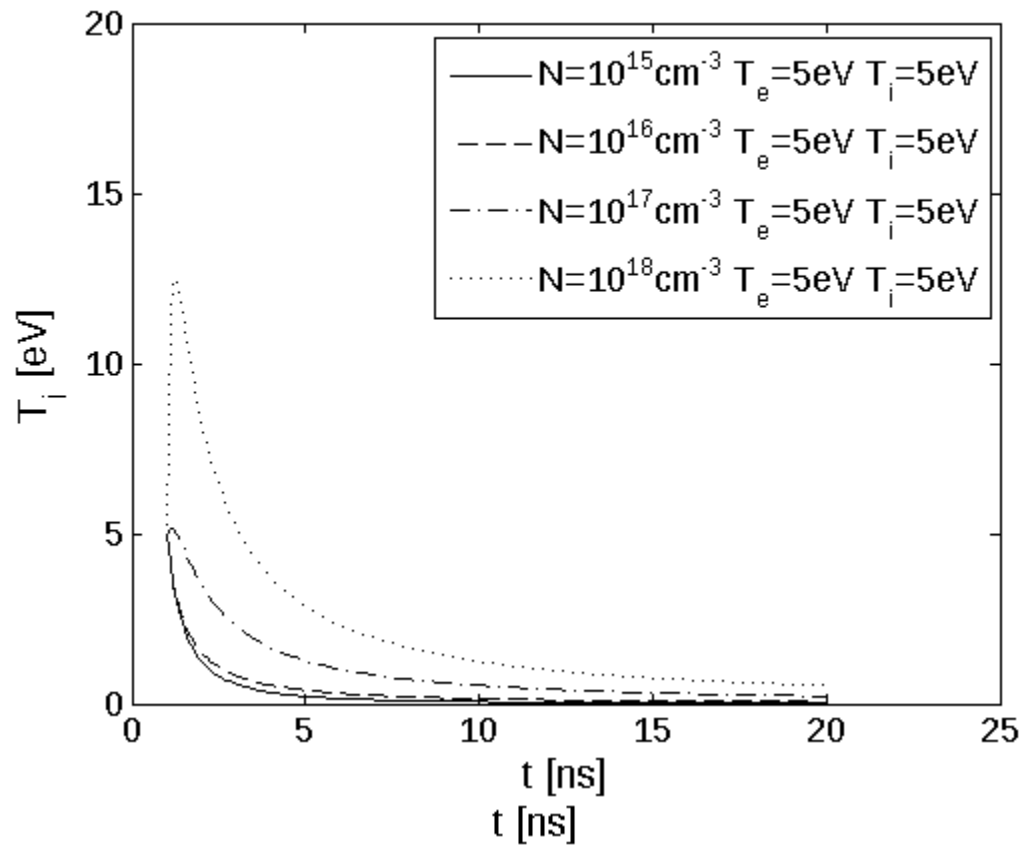


Electron temperature



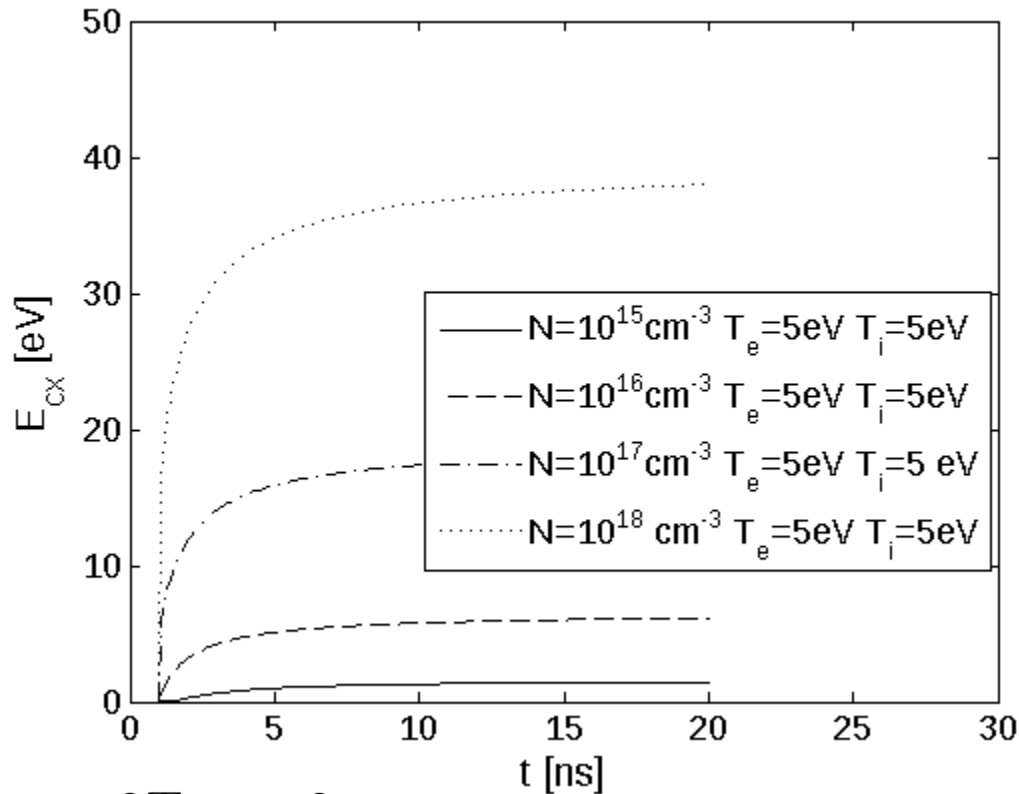


Ion temperature





Energy exchange

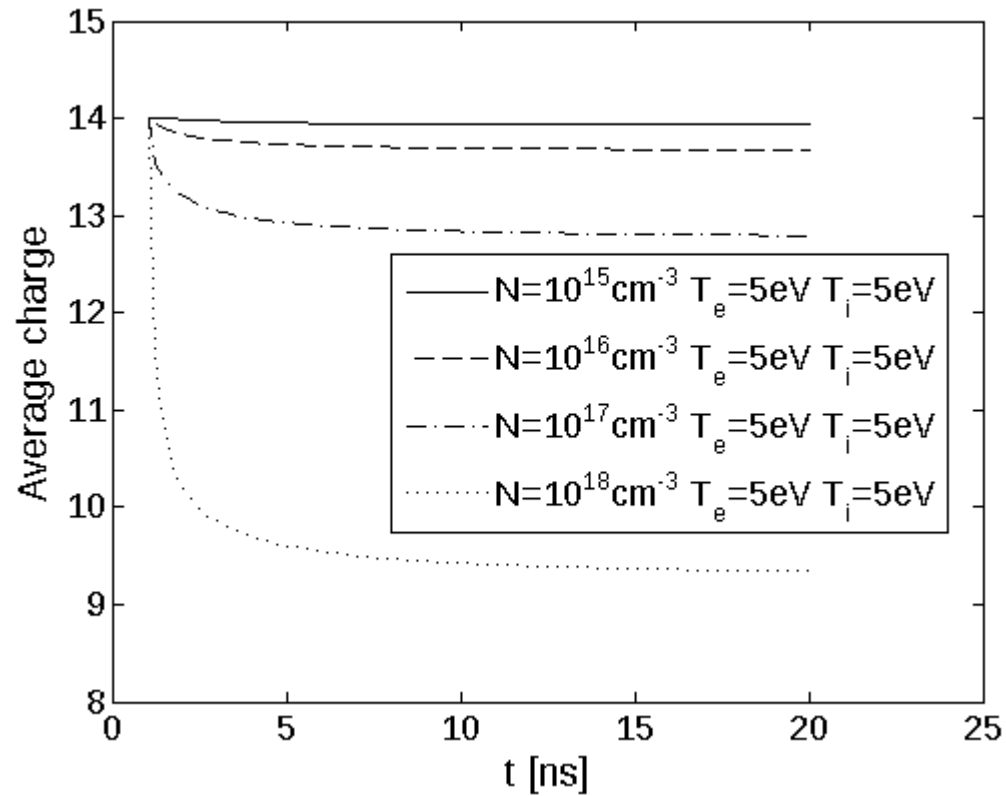


$$\frac{\partial T_e}{\partial t} = \frac{-2T_e}{t} + \frac{2}{3N_e} (P_{\text{recom}} - P_{\text{brem}} - P_{\text{ioniz}}) - P_{ei}$$

$$\frac{\partial T_i}{\partial t} = \frac{-2T_i}{t} + P_{ei} \quad \frac{\partial E_{CX}}{\partial t} = \frac{2}{3N_e} P_{\text{recom}} - P_{ei}$$



Average charge





Conclusion

- The expansion of the plasma after the laser pulses not perfectly adiabatic.
- The three-body recombination acts as an energy source and heats the electrons and consequently changes the electron temperature time dependence.
- Three-body recombination leads to a decrease and freezing of the average charge of the plasma.
- Three-body recombination prolongs the electron-ion relaxation time.
- **OUTLOOK:** Carbon and Aluminum plasmas.

Non-homogeneous expansion



Thanks!

GEFÖRDERT VOM



Bundesministerium
für Bildung
und Forschung



Ronald Redmer

