Nanoplasma dynamics in FEL-driven atomic clusters

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clusters in X-ray pulses: “frustration”  
charge migration: outer & inner tamper  
plasma equilibration: collisional auto-ionization
clusters in X-ray pulses: “frustration”

charge migration: outer & inner tamper

plasma equilibration: collisional auto-ionization
interaction of X-fel pulses with clusters

• **local** interaction:  – inner-shell photo-ionization
  – Auger decays

  intra-atomic processes  **cluster effect?**

• **secondary** processes:  – electron-impact ionization
  – field ionization

• **nanoplasma** dynamics:  – equilibration
  – screening
  – emission

  inter-atomic processes  **cluster effect!**
case study: argon cluster @ X-FEL pulse (350 eV, 80 fs)

[Saalmann & Rost, PRL 89 (2002) 143401]
delocalization of valence states

atomic photo-ionization

\[ \Gamma \propto |\psi(r_\omega)|^2 \quad \text{with} \quad r_\omega = \frac{1}{\sqrt{\omega}} = 0.25 \, a_0 \]

values close to nucleus reduced

auto-ionization (Auger)

\[ \Gamma \propto \left| \langle \Phi_{12}(\vec{x}, \vec{y}) \left| \frac{1}{|\vec{x} - \vec{y}|} \right| \Phi_{0E}(\vec{x}, \vec{y}) \rangle \right|^2 \]

electron-hole overlap reduced
clusters in X-ray laser pulses

cluster-charge effect vs. delocalization effect

[Saalmann & Rost, PRL 89 (2002) 143401]
pulse-length dependence for LCLS pulses

neon @ 2 keV

reduction in clusters for:  
– “long” pulse
– high intensities

absorbed photons (1s) per atom

photons / (100nm)^2
clusters in X-ray pulses: “frustration”
charge migration: outer & inner tamper
plasma equilibration: collisional auto-ionization
plasma formation (for neon clusters and 12 keV X-rays)

- **Auger**: $E_{ex} \approx 0.9$ keV
- **X-ray**: $E_{ex} \approx 11$ keV

- Large radial fields

Cluster size $N$

Cluster radius $R$ [Å]

Average charge $q$

X-ray photo-electron trapping

Auger electron trapping
plasma formation (for neon clusters and 12 keV X-rays)

large radial fields

Bethe rule for atomic (over-barrier) ionization:

$$F_Q \geq \frac{(E_{ip})^2}{4Q}$$

neon: $$F_Q \approx \frac{Q}{7}$$
medium-size neon cluster in strong X-ray pulse
charging of surface atoms \{ 
screening of cluster center \}

\[ \rightarrow \text{consequences for expansion?} \]

quantitative estimate by mean displacement at pulse peak

\[
\Delta r = \frac{1}{N} \sum_{i=1}^{N} |\vec{r}_i(-\infty) - \vec{r}_i(0)|
\]

\[ N \text{ atoms with positions } \vec{r}_i(t) \]

**reduced calculation:**
photo-ionization, Auger decays, electron-impact

**full calculation:**

\[ \ldots, \text{field-ionization due to internal fields} \]

[Gnodtke, Saalmann & Rost, PRA 79 (2009) R 041201]
mean displacement: pulse-length dependence

mean displacement $\Delta r = \frac{1}{N} \sum_{i=1}^{N} |\vec{r}_i(\infty) - \vec{r}_i(0)|$

fixed photon number per pulse: $10^{12}$

full cluster

"half" cluster

$\two\text{H}_2\text{O}$ droplet as sacrificial layer

H$_2$O droplet [Hau-Riege et al. (2007)]
Helium as a tamper

Helium …

… is a weak scatterer (only 2 electrons)

… is transparent to Xray pulses (wouldn’t work without field ionization)

  – immediate response
  – as active as necessary
    ("wächst mit seinen Aufgaben")

… can pick up proteins

Gert von Helden: “Mass-selected protein ions can be stored in an ion trap to be picked up by liquid helium droplets. Detection is performed by direct current measurement.”

… provides an ultra-cold environment
pristine vs. embedded clusters

$\text{Ne}_{1500}$ versus $\text{Ne}_{1500}@\text{He}_{15000}$

pulse-length dependence

Graph showing displacement $\Delta r$ [Å] as a function of pulse duration $T$ [fs] for pristine and embedded clusters.
pristine vs. embedded clusters

pulse-length dependence for $\text{Ne}_{1500}$ versus $\text{Ne}_{1500} @ \text{He}_{15000}$

$$R\text{-factor} = \sum_{ij}^{\text{pixel}} \left| \sqrt{l_{ij}^{\text{real}}} - \sqrt{l_{ij}^{\text{ideal}}} \right| / \sum_{ij}^{\text{pixel}} \sqrt{l_{ij}^{\text{ideal}}}$$

embedding

$\rightarrow$ better images
$\rightarrow$ longer pulse
bio-molecules as heterogenous systems

neon $\leftrightarrow$ helium: different cross sections
electron migration/charge transfer

bio-molecules: carbon, nitrogen, oxygen, sulfur + hydrogen
... and the tamper is on board!

$\rightarrow$ model systems: CH$_4$ (methane) clusters
carbon $\leftrightarrow$ hydrogen: different cross sections
intra-molecular charge transfer

(CH$_4$)$_n$ measurements by Ditmire et al. at LCLS: almost no carbon ions
ultra-fast neutralization

$(\text{CH}_4)_{297}$ @ 1 keV, 10 fs (available at LCLS/Stanford, used by Ditmire et al.)

overall charge within the “carbon core” core is almost neutralized on a femtosecond time scale?
ultra-fast neutralization

$(CH_4)_{297}$ @ 1 keV, 10 fs (available at LCLS/Stanford, used by Ditmire et al.)

- Protons per CH$_4$ molecule within the “carbon core”
- Proton ejection on a femtosecond time scale!

Inverse charge migration $\rightarrow$ inner tamper!
clusters in X-ray pulses: “frustration”
charge migration: outer & inner tamper
plasma equilibration: collisional auto-ionization
xenon clusters at FLASH with 90 eV


\[ E = \hbar \omega - E_{\text{bind}} \approx 20 \text{ eV} \]

4d shell \( E_{\text{bind}} \approx 70 \text{ eV} \)

(large cross section due to xenon’s giant resonance)

- high-energetic tails
- increase with intensity

mechanism?
xenon clusters at FLASH with 90 eV

- laser plasma heating? → no
  - inverse bremsstrahlung decreases strongly with laser frequency $\omega$
  - experiments with $\hbar\omega=40$ eV showed weaker absorption than those with $\hbar\omega=12$ eV

- cluster potential? → no
  - deeper cluster potentials (in particular for higher intensities)

- multi-photon processes? → no
  - low cross sections for direct multi-photon processes
  - low probability for absorption by trapped electrons

→ What else?
simple multi-electron model

placing $N$ electrons in the potential of a homogenous ionic charge distribution

$$V(r) = \begin{cases} 
- \frac{Q}{R} \left[ \frac{3}{2} - \frac{(r/R)^2}{2} \right] & \text{for } r \leq R \quad \text{harmonic with depth } Q/R \\
- \frac{Q}{r} & \text{for } R \leq r \quad \text{Coulombic with charge } Q
\end{cases}$$

“activation” of electrons according to Gaussian pulses

$$I(t) = I_0 \exp \left( -(t/T)^2 \right) \quad \Rightarrow \quad n(t) = (N/2)(1+\text{erf}(t/T))$$

propagation of classical equations of motion

$t \rightarrow -\infty$

$t \approx -T/2$

$t \approx +T/2$
simple multi-electron model

- neglect complicated intra-atomic dynamics, just “activate” electrons with 20 eV excess energy
- account exactly for multi-electron dynamics in the cluster (classical molecular dynamics) and the final “ejection”

parameters

\[
\begin{align*}
R & \quad \text{cluster radius} \\
N & \quad \text{number of activated electrons} \\
T & \quad \text{pulse duration} \\
E^* & \quad \text{excess energy}
\end{align*}
\]

\[ \to \] electron density \( \rho \)
multi-electron dynamics

\[ R = 31 \text{ Å} \quad (\rightarrow \text{Xe}_{2000}) \]

\[ n = 10^4, \, 3 \times 10^3, \, 10^3 \text{ electrons} \]

Gaussian “pulse” \( T = 10 \text{ fs} \)

(charge states for atoms are known from FEL experiments [Richter et al.])

high-energy tails!
time-resolved dynamics

direct:
– sequential (multi-step) ionization
– measured and simulated
[Bostedt et al. PRL 100 (2008) 133401]

plasma:
→ excitation into the cluster potential creating a very dense electron plasma
→ collisional auto-ionization
collisional auto-ionization


\[ \tilde{E} \propto \rho^{1/3} \]

- energy exchange in $e^- - e^-$ collisions
- typical $e^- - e^-$ distances

\[ \frac{1}{r} \propto \rho^{1/3} \]
• clusters in X-ray pulses: “frustration”
  cluster charge, delocalization of valence states

• charge migration: outer & inner tamper
  ultrafast electron migration, proton emission

• plasma equilibration: collisional auto-ionization
  fast electrons in FLASH experiments
  by Bostedt, Möller et al.