Nanoplasma dynamics in FEL-driven atomic clusters

Ulf Saalmann
Max Planck Institute for the Physics of Complex Systems · Dresden

Pierfrancesco Di Cintio · Christian Gnodtke · Ionuţ Georgescu (now at UC Irvine) · Jan M Rost

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

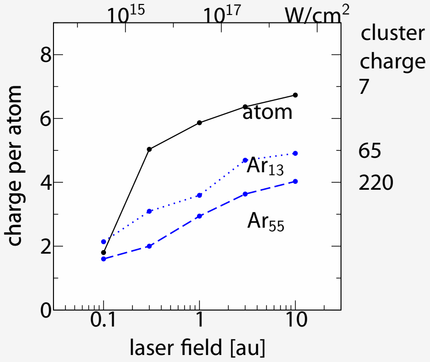
intra-atomic processes **cluster effect ?**

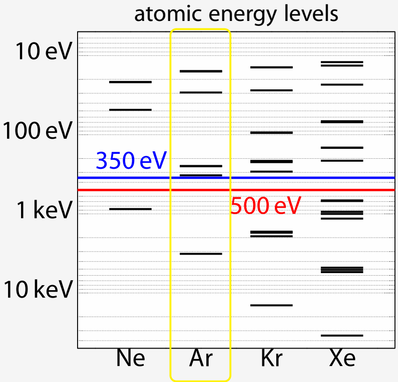
- **secondary** processes: – electron-impact ionization
  – field ionization
- **nanoplasma** dynamics: – equilibration
  – screening
  – emission

inter-atomic processes **cluster effect !**

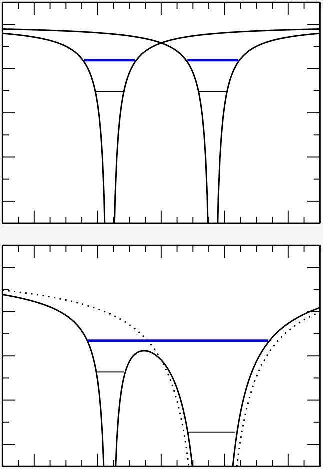
case study: argon cluster @ X-FEL pulse (350 eV, 80 fs)

[Saalmann & Rost, PRL 89 (2002) 143401]





delocalization of valence states



coordinate [au]

atomic photo-ionization

$$\Gamma \propto \left|\Psi(r_\omega)\right|^2 \qquad \text{with} \quad r_\omega = \frac{1}{\sqrt{\omega}} = 0.25\,\mathrm{a}_0$$

values close to nucleus reduced

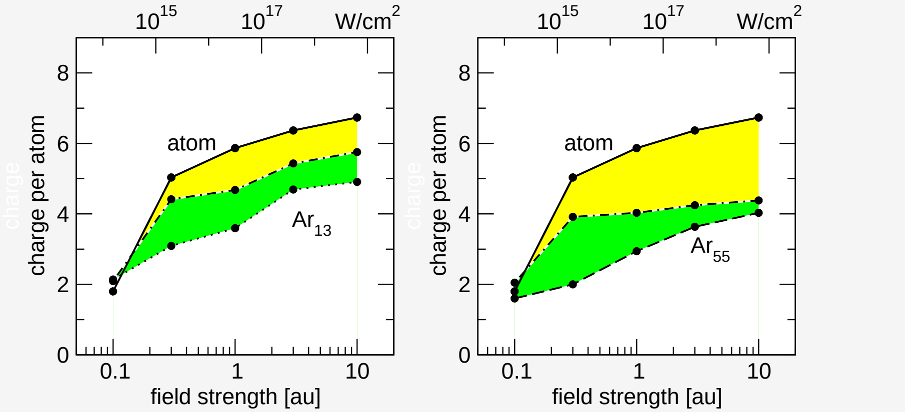
auto-ionization (Auger)

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

electron-hole overlap reduced

cluster-charge effect vs. delocalization effect

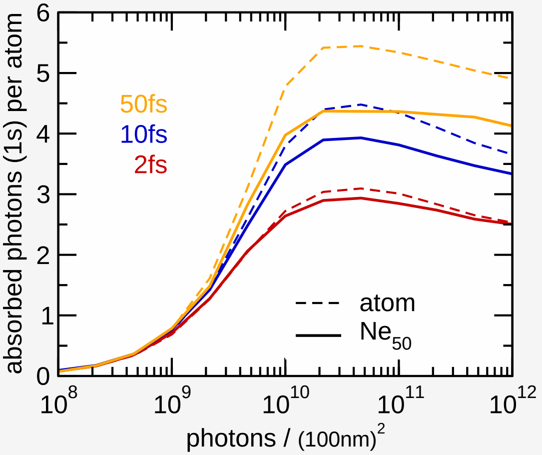
[Saalmann & Rost, PRL 89 (2002) 143401]



pulse-length dependence for LCLS pulses

neon @ 2 keV

reduction in clusters for:
- "long" pulse
- high intensities

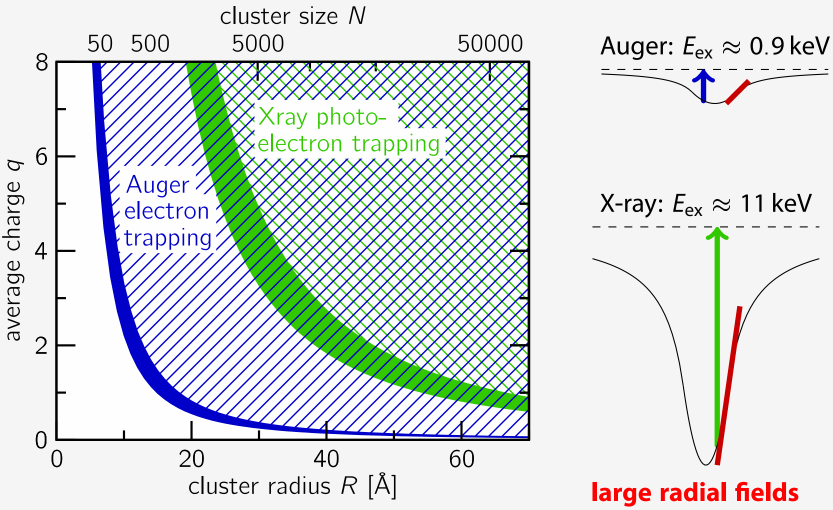


clusters in X-ray pulses: “frustration”

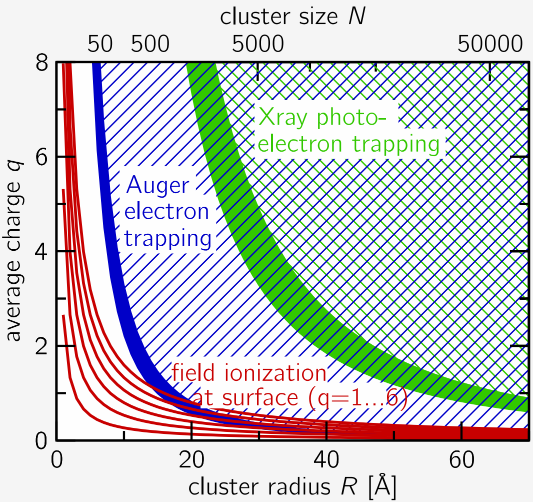
charge migration: outer & inner tamper

plasma equilibration: collisional auto-ionization

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



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

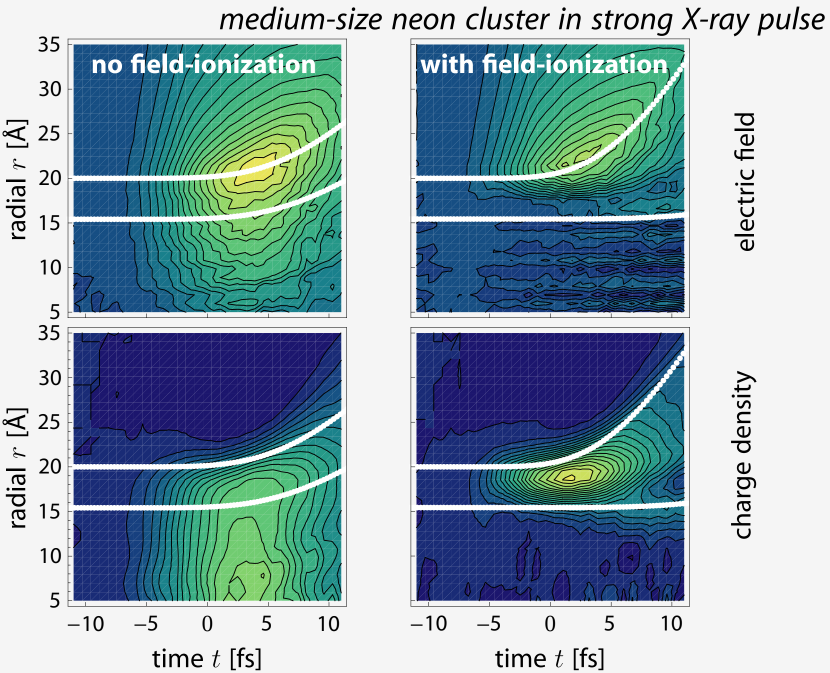


large radial fields

Bethe rule for atomic (over-barrier) ionization:

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

neon: $F_Q \approx \dfrac{Q}{7}$



medium-size neon cluster in strong X-ray pulse
no field-ionization
with field-ionization
electric field
charge density
radial r [Å]
radial r [Å]
time t [fs]
time t [fs]
35
30
25
20
15
10
5
−10
−5
0
5
10

$\left.\begin{array}{r}\text{charging of surface atoms}\\ \text{screening of cluster center}\end{array}\right\} \rightarrow$ 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 atoms with positions $\vec{r}_i(t)$

reduced calculation:

photo-ionization, Auger decays, electron-impact

full calculation:

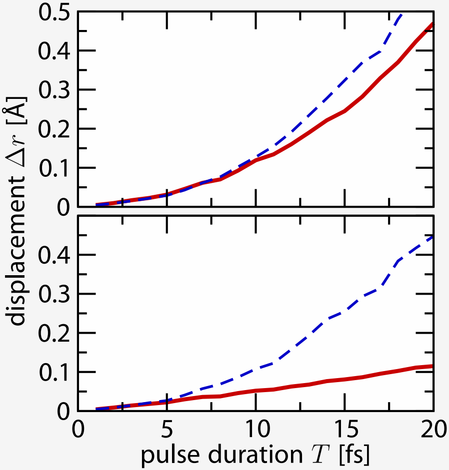
. . . , field-ionization due to internal fields

[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

→ **helium droplet as sacrificial layer**

H_2O 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")

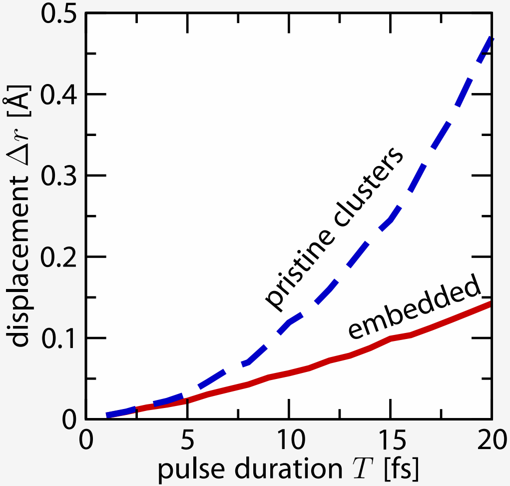
… can pick up proteins

Gert von Helden: (FHI Berlin) "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

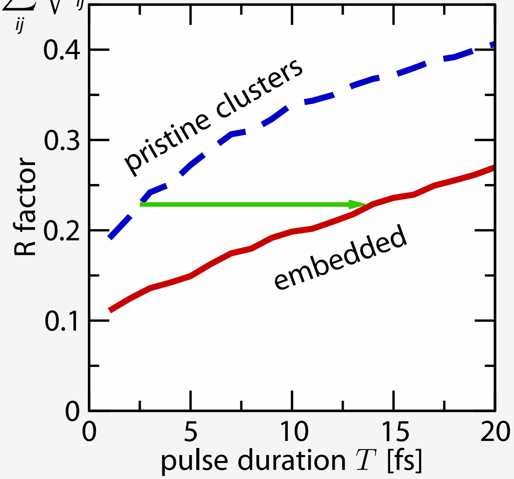
pristine vs. embedded clusters

Ne_{1500} versus Ne_{1500}@He_{15000}
pulse-length dependence



pulse-length dependence for Ne_{1500} versus $Ne_{1500}@He_{15000}$

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



embedding
→ better images
→ longer pulse

bio-molecules as heterogenous systems

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

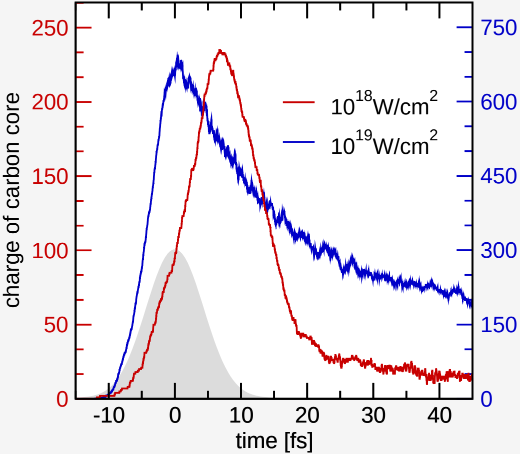
bio-molecules: carbon, nitrogen, oxygen, sulfur **+ hydrogen**
. . . and the tamper is on board !

$\rightarrow$ 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*

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

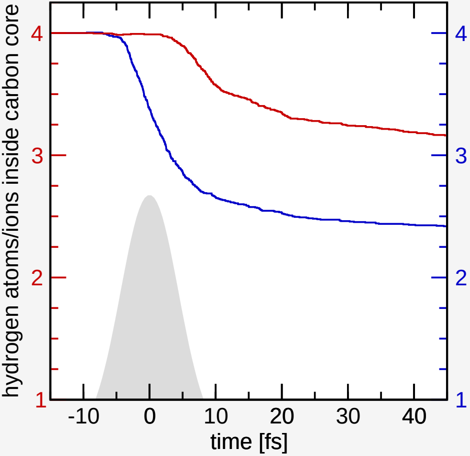


overall charge within the "carbon core"

core is almost neutralized on a femtosecond time scale ?

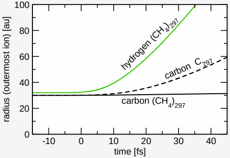
ultra-fast neutralization

$(CH_4)_{297}$ @ 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

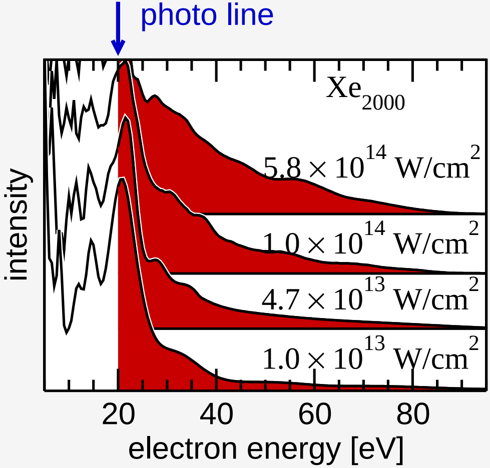
→ inner tamper !

clusters in X-ray pulses: “frustration”

charge migration: outer & inner tamper

plasma equilibration: collisional auto-ionization

xenon clusters at FLASH with 90 eV



[Bostedt, Saalmann et al.
New J. Phys. 12 (2010) 083004]

$E = \hbar\omega - E_{\text{bind}} \approx 20\,\text{eV}$

4d shell xenon: $E_{\text{bind}} \approx 70\,\text{eV}$

(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 ?** $\rightarrow$ **no**
  - inverse bremsstrahlung decreases strongly with laser frequency ω
  - experiments with $\hbar\omega = 40$ eV showed weaker absorption than those with $\hbar\omega = 12$ eV
- **cluster potential ?** $\rightarrow$ **no**
  - deeper cluster potentials (in particular for higher intensities)
- **multi-photon processes ?** $\rightarrow$ **no**
  - low cross sections for direct multi-photon processes
  - low probability for absorption by trapped electrons

$\rightarrow$ **What else ?**

simple multi-electron model

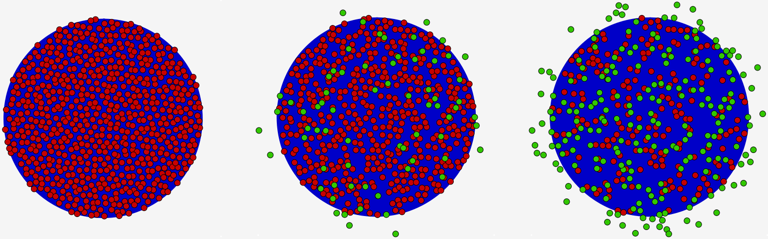
placing N electrons in the potential of a homogenous ionic charge distribution

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

“activation” of electrons according to Gaussian pulses

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

propagation of classical equations of motion



$t \rightarrow -\infty$ $\qquad$ $t \approx -T/2$ $\qquad$ $t \approx +T/2$

- 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

R cluster radius

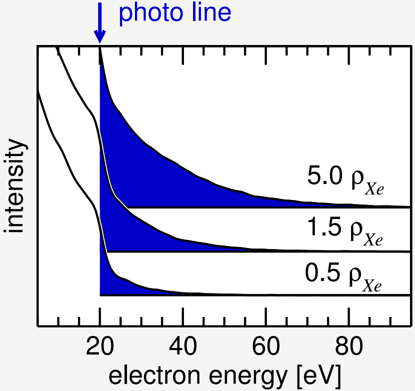
N number of activated electrons

$\left.\begin{array}{l} R \\ N \end{array}\right\} \rightarrow$ electron density ϱ

T pulse duration

$E^\star$ excess energy

simple multi-electron model

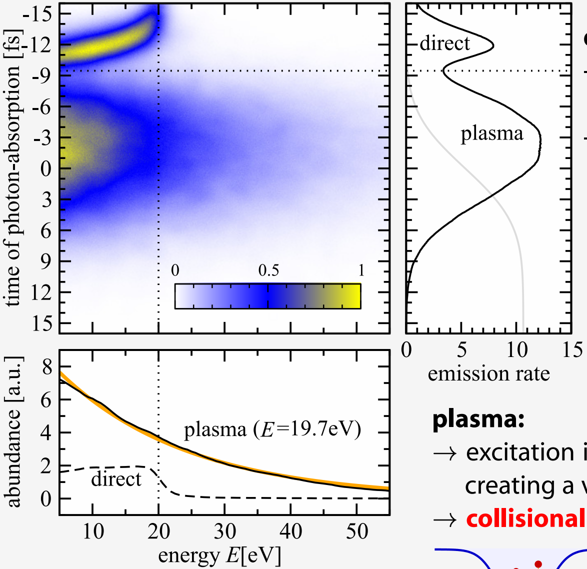


multi-electron dynamics

$R = 31$ Å ($\rightarrow Xe_{2000}$)
$n = 10^4,\ 3\times10^3,\ 10^3$ electrons
Gaussian "pulse" $T = 10$ fs
(charge states for atoms are known from FEL experiments [Richter et al.])

high-energy tails !

time-resolved dynamics



direct:

– sequential (multi-step) ionization

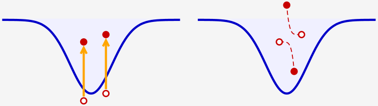
– measured and simulated

[Bostedt et al. PRL 100 (2008) 133401]

plasma:

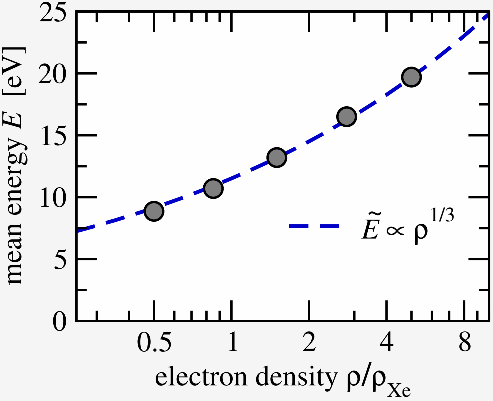
→ excitation into the cluster potential creating a very dense electron plasma

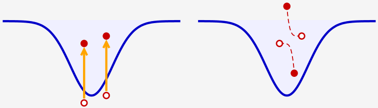
→ **collisional auto-ionization**



collisional auto-ionization

[Bostedt, Saalmann et al. New J. Phys. 12 (2010) 083004]

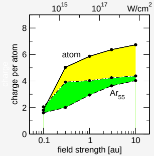




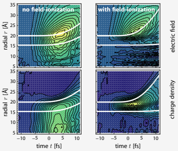
- energy exchange in $e^- - e^-$ collisions
- typical $e^- - e^-$ distances

$$\frac{1}{\bar{r}} \propto \varrho^{1/3}$$

- **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.