Clusters in Intense Laser Pulses: From Infrared to X-Ray Radiation

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**Md. Ranaul Islam**
“Kinetic energy of ions . . .”

**Christian Gnodtke**
“Imaging with X-ray pulses . . .”
- clusters vs. atoms (in strong fields)
- theoretical description
- ionization at optical/infrared pulses
- theory vs. experiment
- time-resolved studies (maybe with attosecond pulses)
- strong X-ray pulses (@ FEL)
(atomic) clusters

Clusters are agglomerates where the number of constituents can be chosen freely.

- Study of properties (e.g., laser-matter interaction) as a function of size
- Different types (metallic, van-der-Waals, etc.); size dependent
- Easily produced (with size distribution); size-selection in collision physics

Bridge between

\[
\begin{align*}
\text{atoms (finite systems)} & \quad \text{but unique properties} \\
\text{solids (high density)} & \quad \text{e.g. very strong charging} \rightarrow \text{explosion}
\end{align*}
\]
clusters in intense laser pulses: experiments

780 nm  “everywhere”
90 nm  Hamburg FEL
3.5 nm  the future

(fast) electrons
(highly-charged, fast) ions
(VUV and X-ray) photons
neutrons
higher charge states and much faster fragments than for molecules!

Ditmire et al. 1997:
- up to Xe\(^{35+}\) with 100 keV
- from Xe\(_{20.000}\)

Ditmire et al. 1999:
- very efficient absorption
- nuclear fusion of fragments

![Graph](image)
platinum: Meiwes-Broer et al. 1999
variable pulse length (fixed fluence)

xenon: Ditmire et al. 1999
pump-probe measurement

dependence on pulse length / pump-probe delay at a femtosecond time scale
\hat{H} = \frac{1}{2} \left(\hat{\vec{p}} - e \vec{A}\right)^2 + V(\vec{r}) \rightarrow \frac{1}{2} \hat{\vec{p}}^2 + V(\vec{r}) - e \frac{\vec{E}}{\omega} \hat{\vec{p}} + \frac{e^2}{2} \frac{\vec{E}^2}{\omega^2} \quad 2E_{\text{pond}} \leftrightarrow E_{\text{bind}}

Keldysh parameter

\gamma \approx \sqrt{\frac{E_{\text{bind}}}{2E_{\text{pond}}}}
= t_{\text{tunnel}} \cdot \omega_{\text{laser}}

\gamma < 1 \quad \text{field ionization}
\quad \text{(over-the-barrier or tunnel)}
clusters vs. atoms in strong fields

\[ V(r) = \begin{cases} 
- \frac{3R^2 - r^2 Q}{2R^2} & r \leq R \\
- \frac{Q}{r} & r \geq R
\end{cases} \]

atom

cluster: \(10^4\) atoms

 expansions

thick barrier

depth \(\sim \frac{Q}{R} \sim R^2\) gradient \(\sim \frac{Q}{R^2} \sim R\)
treatment of laser-cluster interaction

time scales:  
- bound electrons: $10 \ldots 100$ as, $10^{-18}$ s
- laser period (780 nm): $\sim 2$ fs, $10^{-15}$ s
- ionic dynamics: $0.1 \ldots 1$ ps, $10^{-12}$ s
- laser pulse length: $0.1 \ldots 1$ ps, $10^{-12}$ s

two-step approach

- atomic (inner) ionization:
  
  bound electrons $\rightarrow$ “quasi-free” electrons

- cluster (outer) ionization:
  
  “quasi-free” electrons $\rightarrow$ free electrons

Rose-Petruck et al. 1995
atomic (inner) ionization = creation of electrons
  - statistical description by means
    of quantum-mechanical transition rates
    (ADK = field ionization, Lotz = impact ionization)
  - classically in an “onion-like” model:
    new electron if no classically bound electron

cluster (outer) ionization = propagation of electrons (and ions)
  - classical equations of motion
  - by means of a Tree Code for large particle numbers $n$
    because of scaling $\sim n \log n$ (instead of $\sim n^2$)
Cluster-size and pulse-length dependence @ 780 nm

- Average fragment charge:
  - +8
  - +6
  - +4

- Absorbed energy per atom:
  - 32 keV
  - 18 keV
  - 10 keV

Pulses with fixed fluence:

- $2 \times 10^{16}$ W/cm$^2$ @ 100 fs
- $4 \times 10^{15}$ W/cm$^2$ @ 100 fs
pulse-length dependence for different wavelengths

fixed fluence per laser pulse \( (I \cdot T = \text{const}) \)

peak intensity \( I \) [W/cm\(^2\)]

<table>
<thead>
<tr>
<th>pulse length ( T ) [fs]</th>
<th>final charge per atom</th>
</tr>
</thead>
<tbody>
<tr>
<td>10(^{14})</td>
<td>6</td>
</tr>
<tr>
<td>10(^{15})</td>
<td>5</td>
</tr>
<tr>
<td>10(^{16})</td>
<td>4</td>
</tr>
</tbody>
</table>

520 nm, 780 nm, 1170 nm, Xe\(_{561}\)
pulse-length dependence in experiment

platinum clusters [Köller et al, PRL 82 (1999) 3786]
driven damped classical harmonic oscillator

\[ \ddot{X}(t) + 2\Gamma_t \dot{X}(t) + \Omega_t^2 X(t) = F(t) \]

eigen-frequency

\[ \Omega_t = \sqrt{Q_{\text{ion}}/R^3} \]

damping \{ inner ionization, outer ionization \}

periodic driving

\[ F(t) = F_0 \cos(\omega t) \]

\[ X(t) = A_t \cos(\omega t - \phi_t) \]

amplitude

\[ A = F_0/\sqrt{(\Omega_t^2 - \omega^2)^2 + (2\Gamma_t \omega)^2} \]

phase

\[ \phi = \arctan \left( 2\Gamma_t \omega / (\Omega_t^2 - \omega^2) \right) \]

\[ \pi \]

\[ \frac{\pi}{2} \]

0

0

0

0

1

1

2

2

1

2

0

0

1

1

2

2

0

\[ \Omega/\omega \]

\[ \Omega/\omega \]
cluster dynamics \( \text{(Xe}_{923} \ @ \ 9 \times 10^{14} \text{W/cm}^2) \)
\[ \Omega_t^2 = \omega^2 + \left(\frac{F_0}{A_t}\right) \cos \phi_t \]

\[ \Gamma_t = \left(\frac{F_0}{2A_t\omega}\right) \sin \phi_t \]

\[ \Omega_t = \sqrt{\frac{Q_{\text{ion}}(t)}{R(t)^3}} \]
nano-plasma vs. harmonic oscillator model

[Ditmire et al. 1995]

dielectric sphere \( \mathcal{E} = \frac{3}{2 + \varepsilon(\omega)} \mathcal{E}_0 \) with \( \varepsilon(\omega) = 1 - \frac{4\pi \rho}{\omega(\omega + i\nu)} \)

**critical density** \( \omega_{\text{crit}} = \sqrt{\frac{4\pi \rho}{3}} = \frac{\omega_p}{\sqrt{3}} \)

harmonic oscillator model

**eigenfrequency** \( \Omega_t = \sqrt{\frac{Q_{\text{ion}}}{R^3}} = \sqrt{\frac{4\pi \rho_{\text{ion}}}{3}} \)
nano-plasma vs. harmonic oscillator model

**electric field**

**nano-plasma**

Below resonance
\[ \Omega < \omega \]

Above resonance
\[ \Omega > \omega \]

**harmonic oscillator**

negative  \[ \uparrow \]  positive
ionization in a rigid cloud model

Gaussian ion and electron clouds $\rightarrow$ interaction potential

$$\rho(r) = \frac{Q}{\pi^{3/2}R^3} \exp \left( -\frac{(r/R)^2}{2} \right)$$

$$V(x) = \frac{Q}{x} \text{erf}(x/R)$$
ionization in a rigid cloud model

amplitude (harmonic) = barrier (Coulombic)

\[ a = \frac{F}{\Omega^2 - \omega^2} \quad b = \sqrt{\frac{Q}{F}} \]

\[ \rightarrow F(w) = Q^{1/3} \left( \Omega^2 - \omega^2 \right)^{2/3} \]

cycle-averaged potential (KH frame)

\[ \bar{V}(r) = \int_{-\pi}^{+\pi} d\phi \, V(|r - \alpha_0 \sin(\phi)|) \]

becomes a double well

\[ \frac{d^2}{dr^2} \bar{V}(r) \bigg|_{r=0} = 2 \int_0^\pi d\phi \, V''(\alpha_0 \sin(\phi)) < 0 \]

\[ \rightarrow \alpha_0 \approx 2R \quad \text{or} \quad F(w) \approx 2R\omega^2 \]
ionization in a many-electron model

ionization probability

$2^{13} = 8192$ electrons in a jellium potential

PSfrag replacements

ionization probability
cluster-size and pulse-length dependence @ 780 nm

average fragment charge

absorbed energy per atom

pulses with fixed fluence

2 \times 10^{16} \text{ W/cm}^2 @ 100 \text{ fs}

4 \times 10^{15} \text{ W/cm}^2 @ 100 \text{ fs}
ionization in ultra-short pulses

cluster-size dependence for $T = 25\text{ fs}$

at $I = \frac{16}{3.2} \times 10^{15} \text{ W/cm}^2$

field ionization model

$q \propto \sqrt{I/R} \quad E \propto I$

average fragment charges

absorbed energy per atom
kinetic energy distribution of ions

“standard” experimental observable
data from different groups studying different targets

Ditmitre et al. (1997)

\[ \text{Xe}_{2500} \]

\[ \begin{align*}
E\ [\text{keV}] & \quad 10^0 & 10^1 & 10^2 & 10^3 \\
\text{KEDI [arbitrary units]} & \quad 10^5 & 10^4 & 10^3 & 10^2 & 10^1 & 10^0
\end{align*} \]

Ditmitre et al. (2001)

\[ \text{Xe}_{9000} \]

\[ \begin{align*}
E\ [\text{keV}] & \quad 10^1 & 10^2 \\
\text{KEDI [arbitrary units]} & \quad 10^3 & 10^2 & 10^1 \quad 10^0
\end{align*} \]

Mathur et al. (2003)

\[ \text{Ar}_{40000} \]

\[ \begin{align*}
E\ [\text{keV}] & \quad 10^1 & 10^2 \\
\text{KEDI [arbitrary units]} & \quad 10^3 \quad 10^2 \quad 10^1 \quad 10^0
\end{align*} \]
kinetic energy distribution of ions

microscopic single-cluster calculations for Xe$_{9093}$

- $1.6 \times 10^{16}$ W/cm$^2$ @ 25 fs
- $4 \times 10^{15}$ W/cm$^2$ @ 100 fs
- $8 \times 10^{14}$ W/cm$^2$ @ 400 fs
kinetic energy distribution of ions

microscopic single-cluster calculations for Xe\textsubscript{9093}

1.6 \times 10^{16}\text{W/cm}^2 @ 25\text{fs}  \hspace{1cm} 4 \times 10^{15}\text{W/cm}^2 @ 100\text{fs}  \hspace{1cm} 8 \times 10^{14}\text{W/cm}^2 @ 400\text{fs}
simple model of a expanding charged sphere
shell at radial distance $r$: energy $E \propto r^4$ $\rightarrow$ field $F \propto r^3$ $\rightarrow$ acceleration $a \propto r$
$\rightarrow$ cluster keeps its shape
kinetic energy distribution of ions

single cluster in a homogeneous field

homogeneous charge distribution in the cluster

\[ \frac{dP}{d\varepsilon} = \frac{3}{2} \sqrt{\varepsilon} \Theta(1 - \varepsilon) \]

fastest ions from the surface

Note: scaled energies!
kinetic energy distribution of ions

clusters from Gaussian laser focus

\[ \sim \exp \left( -\frac{r^2}{\xi^2} \right) \]

\[
\frac{dP_{\text{las}}}{d\varepsilon} = \frac{\pi\xi^2 N}{2} \frac{1 - \varepsilon^{3/2}}{\varepsilon} \Theta(1 - \varepsilon)
\]

higher charges for clusters in the focus than those in the tails
kinetic energy distribution of ions

cluster size distribution in a fixed field

folding with cluster size distribution

\[ \sim \frac{1}{N} \exp \left( -\frac{\ln^2(N/N_0)}{2\nu^2} \right) \]

\[ \frac{dP_{\text{size}}}{d\varepsilon} = \frac{3}{4} N_0 \sqrt{\varepsilon} \text{erfc} \left( \frac{3}{2} \frac{\ln \varepsilon}{\sqrt{2\nu}} \right) \]

fastest ions from larger clusters
kinetic energy distribution of ions

cluster size distribution from Gaussian laser

folding with both distributions

\[
\frac{dP_{\text{both}}}{d\varepsilon} = \frac{\xi^2 \pi}{4} \frac{N_0}{\varepsilon} \left[ \varepsilon^{3/2} \text{erfc} \left( \frac{3 \ln \varepsilon}{2 \sqrt{2\nu}} \right) + e^{\nu^2/2} \left( 1 + \text{erf} \left( \frac{2\nu^2 - 3 \ln \varepsilon}{2 \sqrt{2\nu}} \right) \right) \right]
\]

separate modifications of the spectrum at its two different “ends”!
kinetic energy distribution of ions

saturation effect: simple relation $q(r) \sim \mathcal{E}(r)$ may break down for large $\mathcal{E}$, i.e. small $r$

because of finite number of electrons available (e.g. hydrogen: 1, N$_2$: 5 per atom)

$$\frac{dP_{\text{sat}}(\eta)}{d\varepsilon} = \frac{dP_{\text{both}}}{d\varepsilon} - \ln \eta \frac{dP_{\text{size}}}{d\varepsilon}$$
kinetic energy distribution of ions

model vs. experimental data for \((N_2)_N\) clusters of different sizes \(N\)

\[
\frac{E_{\text{sat}}}{N_0} = \frac{R}{N_0^2} = 3
\]
kinetic energy distribution of ions


if common saturation charge for all cluster sizes

\[ E_{\text{sat}} \propto N_0/R \propto N_0^{2/3} \]
kinetic energy distribution of ions

experimental data from different groups studying different targets
ionization at shorter wavelengths / higher frequencies

780 nm, 1.5 eV  
\( \gamma \ll 1 \)  
\( \sim \) cluster size

90 nm, 13 eV  
\( \gamma \sim 1 \)  
\( \sim \) atom size

3 nm, 350 eV  
\( \gamma \gg 1 \)  
\( \sim 0 \)

Keldysh parameter
quiver amplitude

inner ionization
barrier suppression

outer ionization
collective heating

single-photon absorption (into cluster)
inverse bremsstrahlung

single-photon absorption or auto-ionization (into continuum)

treatment of laser-cluster interaction

**time scales:**
- bound electrons: $10 \ldots 100 \text{ as} = 10^{-18} \text{ s}$
- laser period (3 nm): $\sim 10 \text{ as} = 10^{-18} \text{ s}$
- ionization rates: $0.1 \ldots 10 \text{ fs} = 10^{-15} \text{ s}$
- ionic dynamics: $0.1 \ldots 1 \text{ ps} = 10^{-12} \text{ s}$
- laser pulse length: $0.1 \ldots 1 \text{ ps} = 10^{-12} \text{ s}$

**mixed quantenmechanical–classical approach**

- atomic ionization and intra-atomic decays
  - statistical description by means of *quantenmechanical* transition rates (photo-ionization, auto-ionization)
- dynamics of free electrons and ions
  - propagation of *classical* equations of motion
X-FEL: ionization of atoms “inside–out”

(1) **inner-shell photo-ionisation**

\[ \Gamma = I [\text{au}] \cdot 0.1 \text{ fs}^{-1} \]

(2) decay cascades:

- **Auger decays** \[ \Gamma_{\text{Argon}} = 0.2 \ldots 5 \text{ fs}^{-1} \]
- radiative transitions
- “shake-off” processes

expectation: multiple photo-ionization fast decay cascades during pulse → enormous energy absorption
argon cluster @ X-FEL pulse (350 eV, 80 fs)

charge per atom vs. laser field [au]

atomic energy levels

Ne  Ar  Kr  Xe

ionization potentials

10 eV  100 eV  1 keV  10 keV  500 eV  350 eV
argon cluster @ X-FEL pulse (350 eV, 80 fs)

large space charge & small quiver amplitude \( \sim \frac{\sqrt{I}}{\omega^2} \)

delocalization of valence states
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

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

overlap of excited electrons and hole reduced

coordinate [au]
pulse (350 eV, 1 au): ionized electrons

photo-ionization  tunneling  photo- and auto-ionization

![Graph of ionization for different Ar isotopes](image)
pulse (350 eV, 1 au): ionized electrons without tunneling

photo-ionization  tunneling  photo- and auto-ionization

\[ \text{Ar}_{13} \]

\[ \text{Ar}_{55} \]
clusters in X-ray laser pulses

space-charge effect vs. delocalization effect

[US & Rost, PRL 89 (2002) 143401]
The End!