Clusters in intense laser fields

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Outline

— **Introduction**
  
  — Experimental facts
  — Frequently used concepts

— **Laser-cluster interaction via semi-classical molecular dynamics simulations with explicit treatment of inner-atomic dynamics**

  ... in order to see what’s really going on

  — Ionization mechanism
  — Energy absorption at long and short wavelengths

— **Harmonic generation via nonlinear Mie-plasmon excitation**

— **Summary**
Introduction

Why is it of interest to shoot with a laser on clusters?

- **Efficient coupling of laser energy into matter** because
  - density is (locally) high (≠ gas targets)
  - yet the cluster size is smaller than the laser wavelength and skin depth
  - energy remains confined (no cold bulk)

→ **High charge states** (much higher than expected from field ionization by the laser alone)
→ **Energetic ions** (MeV), electrons (keV), and photons (x-rays)
→ **Fusion in deuterium clusters**
Experimental facts

Ions:

Figure 2 Ion energy spectrum from 65 Å (~2,500 atom) Xe clusters irradiated by a peak intensity of $2 \times 10^{16} \text{ W/cm}^2$, derived from the ion TOF trace shown as an inset. (The first peak in the TOF trace corresponds to fast (>2-keV) electrons; the feature at later times corresponds to the ions. Note that electrons with energy <2 keV do not reach the MCP detector as the front plate is charged to ~2,000 V).

Figure 3 Measured charge-state distribution of 65 Å Xe clusters irradiated by a peak laser intensity of $2 \times 10^{16} \text{ W/cm}^2$ for ion kinetic energies of 1, 3, 10, 30 and 100 keV.

Ditmire et al., Nature 386, 54 (1997)

$2 \times 10^{16} \text{ W/cm}^2$, Xe cluster, $2R \approx 65\text{Å}$ ($\rightarrow$ 2500 atoms/cluster)

Isolated Xe, Bethe rule $E = \varepsilon^2/4Z$: only Xe$^{8+}$ expected
Experimental facts

Photons:

X-rays in the 2–3 Å-regime in 300 fs, $10^{18}$ W/cm$^2$ laser-cluster (Xe) experiments ($\rightarrow$ charge states Xe$^{45+}$ involved, “hollow atoms”)


$\approx 10^{18}$ W/cm$^2$, 248 nm


$\approx 2 \times 10^{18}$ W/cm$^2$, 800 nm

Ter-Avetisyan et al., Phys. Rev. E 64, 036404 (2001)
H. Wabnitz et al., Nature 420, 482 (2002)
98 nm (12.7 eV), 100 fs,
up to $7 \times 10^{13}$ W/cm$^2$

**letters to nature**

**Multiple ionization of atom clusters by intense soft X-rays from a free-electron laser**


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|| Joint Institute for Nuclear Research, Dubna, 141980 Moscow Region, Russia

**Figure 1** Time-of-flight (TOF) mass spectra of ionization products of Xe atoms and clusters. The spectra were recorded after ionization with soft X-rays (98 nm wavelength) at an average power density of $2 \times 10^{13}$ W cm$^{-2}$. The atomic spectrum (bottom trace) shows a splitting into several lines owing to the different isotopes. After irradiation of clusters, highly charged ions are observed. The mass peaks are rather broad and displaced with respect to the calculated flight times indicated by thin vertical lines (different charge states) in the uppermost part of the figure. This indicates that the ions have high kinetic energies. $N$ is the number of atoms per cluster. Inset, the kinetic energy of ions as a function of the charge for $N = 1,500$. The experiments are performed as follows. A pulsed beam of atoms or clusters was prepared by expanding Xe gas at high pressure (0.1–30 bar) through a small conical nozzle (0.1 mm diameter, 15° half-angle). The composition of the beam and the size of the clusters could be controlled by varying the gas pressure. The cluster beam passed through a small skimmer into the main chamber. Soft X-rays from the FEL with 98 nm wavelength (bandwidth 0.5 nm) were focused with an elliptical mirror onto the atom or cluster beam. The pulse energy was measured with a carefully calibrated channel-plate detector, which detects a well-defined percentage of FEL light scattered from a wire. The diameter of the focal spot was ~20 μm. The power density is calculated assuming a pulse length of 100 fs. The ions produced in the interaction zone were detected with a TOF detector mounted perpendicular to the polarization direction and with a digital sampling oscilloscope operating at a 1-Hz repetition rate.
Why high charge states? — Ionization ignition


Inner ionization

Electrons leave their parent ions

Outer ionization

Some electrons leave the cluster

Potential $\approx$ harmonic inside
$\rightarrow$ Mie frequency $\omega_{\text{Mie}}^2 = \omega_p^2 / 3 = 4\pi Z N_a / 3 V_c$
Ionization ignition

$F_{\text{cluster}}$ leads to increased inner ionization

Example: Cluster of 1000 Xe atoms, all doubly ionized $\Rightarrow 4 \times 10^{11}$ V/m at the surface, corresponding to $2 \times 10^{16}$ W/cm$^2$

... Coulomb explosion, energetic particles

but ...

ionization ignition breaks down soon if the electrons stay inside the cluster

Mechanism for efficient outer ionization needed!
MD simulations of clusters in laser fields I

A few references (not exhaustive!):

→ ionization ignition

→ dependence of ionization on laser frequency and cluster radius

→ resonance enhancement of outer ionization

→ Coulomb explosion rather than hydrodynamic expansion, ionization ignition rather than collisional ionization

→ nuclear fusion in Coulomb-exploding (D$_2$O)$_n$ clusters
MD simulations of clusters in laser fields II


Christian Siedschlag and Jan M. Rost, physics/0310123v2 (2003) → clusters in VUV fields: ionization threshold reduction, inverse bremsstrahlung

Semi-classical (fermionic) molecular dynamics SC(F)MD

**Problem:** Simulating the interaction of rare gas clusters with intense laser fields on a TDDFT level is too demanding in 3D

**“Solution”:** Classical or semi-classical simulations

**Problem:** Classical many-electron atoms are unstable

**“Solution”:** Treat inner ionization via ensembles of (hydrogen-like) Kepler orbits (CTMC) or through rate equations

(Rose-Petruck *et al.*, 1997; Last and Jortner, 1999, 2000; Saalmann and Rost, 2002; Siedschlag and Rost, 2002, 2003; ...)

**Our approach:** SC(F)MD makes classical multi-electron atoms stable so that *inner ionization can be treated self-consistently*

Kirschbaum-Wilets approach

Momentum-dependent potential

\[ V(r, p, \xi, \alpha, \mu) = \frac{\xi^2}{4\alpha r^2 \mu} \exp \left\{ \alpha \left[ 1 - \left( \frac{rp}{\xi} \right)^4 \right] \right\} \]

enforcing \( rp \geq \xi \) more (big \( \alpha \)) or less (small \( \alpha \)) severely; \( \mu \): reduced mass

Heisenberg uncertainty relation

\[ V_H(\tilde{r}_{ij}, \tilde{p}_{ij}) = V(\tilde{r}_{ij}, \tilde{p}_{ij}, \xi_H, \alpha_H, 1) \]

where \( \tilde{r}_{ij} = |r_i - R_j| \) and \( \tilde{p}_{ij} = |(Mp_i - P_j)/(1 + M)| \)

Pauli principle \( \to \) shell structure

\[ V_P(\tilde{r}_{jl}, \tilde{p}_{jl}, \sigma_j, \sigma_l) = V(\tilde{r}_{jl}, \tilde{p}_{jl}, \xi_P, \alpha_P, 1/2) \delta_{\sigma_j \sigma_l} \]

where \( \tilde{r}_{jl} = |r_j - r_l| \) and \( \tilde{p}_{jl} = |p_j - p_l|/2 \)
Semi-classical molecular dynamics simulations of clusters in laser fields with explicit inner-atomic dynamics

$N_a$ ions, $Z$ “active” electrons per ion, laser field $E(t)$

$$H(R, P; r, p; t) = \sum_{i=1}^{Na} \frac{P_i^2}{2M} + \sum_{j=1}^{Na} \frac{P_j^2}{2}$$

$$+ \sum_{i=1}^{Na} \sum_{j=1}^{Na} \left( V_H - \frac{Z}{|R_i - r_j|} \right)$$

$$+ \sum_{i=1}^{Na} \sum_{k=1}^{i-1} \frac{Z^2}{|R_i - R_k|}$$

$$+ \sum_{j=1}^{Na} \sum_{l=1}^{j-1} \left( V_P + \frac{1}{|r_j - r_l|} \right)$$

$$+ E(t) \cdot \left( \sum_{j=1}^{Na} r_j - Z \sum_{i=1}^{Na} R_i \right)$$

“Heisenberg-potential” $V_H$

“Pauli-potential” $V_P$

D.B., J. Phys. B (submitted)
The single, isolated atom

5p shell of Xe, $\xi_H = 2.33$, $\alpha_H = 2$, $Z = 6$

<table>
<thead>
<tr>
<th>charge state</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
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</thead>
<tbody>
<tr>
<td>real $E_I$</td>
<td>0.45</td>
<td>0.77</td>
<td>1.18</td>
<td>1.69</td>
<td>2.09</td>
<td>2.64</td>
</tr>
<tr>
<td>model $E_I$</td>
<td>0.16</td>
<td>0.55</td>
<td>1.11</td>
<td>1.63</td>
<td>2.22</td>
<td>2.65</td>
</tr>
</tbody>
</table>

Results for the Xe\(_{54}\) model cluster

\(N_a = 54, \ Z = 6\) (the six 5p-electrons), \(r_W = 4\) a.u., \(R_0 \approx 15.4\) a.u.

Tabulated configurations for \(N_a < 150\): [http://brian.ch.cam.ac.uk/](http://brian.ch.cam.ac.uk/)

Laser parameters: \(\lambda_l = 800\) nm, trapezoidal \((3,10,3)\)-pulse, \(\lambda_s = 100\) nm, \((24,80,24)\)-pulse, \(\rightarrow T \approx 42\) fs

Three groups of electrons, contributing to:

— “parent atom ionization”: \(N_{\text{parent}} = \frac{1}{N_a} \sum_{i=1}^{N_a} \Theta(|r_j - R_{\text{parent}}| - r_{WS})\)

— average charge state: \(Z_{\text{avcs}} = \frac{1}{N_a} \left( ZN_a - \sum_{i=1}^{N_a} \sum_{j=1}^{ZN_a} \Theta(r_{WS} - |r_j - R_i|) \right)\)

— outer ionization: \(N_{\text{outer}} = \frac{1}{N_a} \sum_{j=1}^{ZN_a} \Theta(\max\{R_i\} + r_{WS} - r_j)\)
Inner, outer, and “parent atom” ionization

\[ I = \hat{E}^2 = 6.4 \times 10^{-3} \implies 2.2 \times 10^{14} \text{ W/cm}^2, \ T \approx 42 \text{ fs} \]
Relative ionization at 800 and 100 nm

![Graph showing relative charge and laser intensity relationship.](image)
Fields inside the cluster at 800 nm

Fields inside the cluster at 100 nm

 Ionization ignition after thermionic emission of electrons
Charge state distributions

Ignition field $\rightarrow$ charge states inside the cluster increase with the ion radii.

$\rightarrow$ broad charge state distributions

Example: $I = 6.4 \times 10^{-3} \ (2.2 \times 10^{14} \text{ W/cm}^2)$
Energy absorption

Absorbed energy

\[ \Delta \mathcal{E} = \mathcal{H}(T) - \mathcal{H}(0) = \sum_{i=1}^{N_a} \Delta \mathcal{E}_i + \sum_{j=1}^{ZN_a} \Delta \mathcal{E}_j = \int_0^T E(t) \cdot J \, dt \]

\[ J = Z \sum_i \dot{R}_i - \sum_j \dot{r}_j, \quad \Delta \mathcal{E}_i = Z \int_0^T E(t) \cdot \dot{R}_i \, dt, \quad \Delta \mathcal{E}_j = - \int_0^T E(t) \cdot \dot{r}_j \, dt \]

For absorption: Phase lag between current \( J \) and field \( E \) \( \neq \pm \pi/2 \)

Possible through

— electron-ion collisions (inverse bremsstrahlung)
— electron-cluster boundary collisions
— electron-cluster collisions
Total and “outer” energy absorption

\[
\Delta E_{oa} = - \int_0^T \mathbf{E}(t) \cdot \sum_{j=1}^{ZN_a} \mathbf{r}_j \prod_{i=1}^{Na} \Theta(|\mathbf{r}_j - \mathbf{R}_i| - r_{WS}) \, dt
\]
How do electrons absorb laser energy?

Colors indicate cycle-averaged absorption rate

$$\dot{E}_j(t) = -\frac{1}{T} \int_{t-T/2}^{t+T/2} E(t') \cdot \dot{r}_j dt'$$

Low: dark colors; high: light colors
Third harmonic generation via Mie-plasmon excitation

Idea: During cluster expansion the resonance condition

\[ \omega_{\text{Mie}} = \sqrt{\frac{4\pi Z N_a}{3V_c}} = 3\omega \]

is reached before the “linear resonance” \( \omega_{\text{Mie}} = \omega \).

Preliminary experimental results from pump-probe measurements have been obtained (G. R. Hays et al., SILAP Conference 2003, Dallas, TX)

Example: \( \text{Xe}_{720}, \tilde{E} = 0.5 \left(8.775 \times 10^{15} \text{ W/cm}^2\right), T = 3307 \ (79 \text{ fs}, 30 \text{ cycles}), \omega = 0.057 \ (800 \text{ nm}) \)
### Spectrum and time profiles

$Xe_{720}, \hat{E} = 0.5 \ (8.775 \times 10^{15} \text{ W/cm}^2), \ T = 3307 \ (79 \text{ fs, 30 cycles}), \ \omega = 0.057 \ (800 \text{ nm})$

<table>
<thead>
<tr>
<th>Harmonic order</th>
<th>Emitted power (arb.u.)</th>
<th>Time (a.u.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2w</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3w</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4w</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

![Graph showing emitted power and time profiles](image-url)
— Resonant third harmonic generation confirmed in simulation and experiment

**So what?**

— The nonlinear excitation of the Mie-plasmon has seemingly no strong effect on energy absorption and ionization

**But ...**

— Useful as a diagnostics tool

— Width of the harmonic emission vs the laser wavelength yields information about damping (Landau and collisional)
Summary

• Cluster efficiently absorb laser energy $\rightarrow$ high charge states, fast ions and electrons, x-rays, fusion neutrons.

• Ionization in small clusters ($N_a < 10^3$) is due to
  — dynamical ionization ignition (at long wavelengths)
  — thermionic electron emission + ionization ignition (at short wavelengths)

• Laser energy absorption proceeds through
  — electron-cluster (boundary) collisions (at long wavelengths)
  — inverse bremsstrahlung (at short wavelengths)

• Nonlinear, resonant excitation of the Mie-plasmon (accompanied by third harmonic-emission) was observed in simulations and pump-probe experiments
Experimental facts???

Electrons:

![Graph](image1.png)

**FIG. 1.** (a) Measured electron kinetic energy distribution from Xe clusters for a peak intensity of $1 \times 10^{19}$ W/cm$^2$. The gas jet backing pressure was 4.5 bars (corresponding to an average cluster size of 50 Å). (b) Calculated electron energy distribution for a peak intensity of $1 \times 10^{19}$ W/cm$^2$ and a cluster size of 50 Å.


Hot electron peak not reproducible!

![Graph](image2.png)


Experimental facts

Neutrons:

Deuterium clusters in 35 fs, 820 nm, $\approx 2 \times 10^{16}$ W/cm$^2$ laser pulses $\rightarrow$ keV ions


\[
D + D \rightarrow He^3 + n \ (2.45 \text{ MeV})
\]

$10^5$ fusion neutrons per joule of incident laser energy
A different point-of-view: The nanoplasma model


— Outer electrons are removed from their parent ions by the laser

— Absorption of laser energy by electron-ion collisions

— Collisional ionization

— Outer ionization small → quasi-neutral plasma

— Adiabatic hydrodynamic expansion

— Screening or field-enhancement, Mie-resonance; Drude model:

\[ E = \Re\{\hat{E}\exp(i\omega t)\}, \quad E_{\text{inside}} = \Re\left\{ \frac{3\hat{E}}{\epsilon + 2}\exp(i\omega t) \right\}, \quad \epsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\nu_{ei})} \]

for \( \omega = \omega_p/\sqrt{3} \Rightarrow \hat{E}_{\text{inside}} > \hat{E} \Rightarrow \) enhanced absorption and ionization
Nanoplasma model vs experiment

The good point: cluster expansion and Mie-resonance → optimal cluster size and pulse length


Pulse shape dependence: Mendham et al., Optics Express 11, 1357 (2003)
Nanoplasma model vs experiment

The not so good points: wavelength dependence, resonance time, quantitative agreement


Improved plasma models:

Non-uniform cluster plasma → long-time resonance.

Milchberg et al., Phys. Rev. E 64, 056402 (2001)
Dynamical ionization ignition

TDDFT
1D model
800 nm
\( \hat{E} = 0.033 \)
(3, 8, 3)-pulse