Ionization dynamics of clusters in laser fields

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Outline

— Introduction
  – Why clusters?
  – Ionization ignition

— 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

— 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
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 $\omega_M^2 = \omega_p^2/3 = 4\pi Z N_a/3 V_c$
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!
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 \( \tilde{p}_{ij} = |(M p_i - P_j)/(1 + M)| \)

Pauli principle \( \rightarrow \) 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 $E(t)$

\[
\mathcal{H}(\mathbf{R}, \mathbf{P}; \mathbf{r}, \mathbf{p}; t) = \sum_{i=1}^{N_a} \frac{\mathbf{P}_i^2}{2M} + \sum_{j=1}^{ZN_a} \frac{\mathbf{p}_j^2}{2} 
+ \sum_{i=1}^{N_a} \sum_{j=1}^{ZN_a} \left( V_H - \frac{Z}{|\mathbf{R}_i - \mathbf{r}_j|} \right) 
+ \sum_{i=1}^{N_a} \sum_{k=1}^{i-1} \frac{Z^2}{|\mathbf{R}_i - \mathbf{R}_k|} 
+ \sum_{j=1}^{ZN_a} \sum_{l=1}^{j-1} \left( V_P + \frac{1}{|\mathbf{r}_j - \mathbf{r}_l|} \right) 
+ \mathbf{E}(t) \cdot \left( \sum_{j=1}^{ZN_a} \mathbf{r}_j - Z \sum_{i=1}^{N_a} \mathbf{R}_i \right)
\]

“Heisenberg-potential” $V_H$

“Pauli-potential” $V_P$
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/

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 vs. laser intensity for charge state and outer layers.](image)
Fields inside the cluster at 800 nm

Dynamical ionization ignition

Charge state distributions

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

$\implies$ broad charge state distributions

Example: $I = 6.4 \times 10^{-3} \ (2.2 \times 10^{14} \text{ W/cm}^2)$
How do electrons absorb laser energy?

Colors indicate cycle-averaged absorption rate

\[ \dot{\mathcal{E}}_j(t) = -\frac{1}{T} \int_{t-T/2}^{t+T/2} \mathbf{E}(t') \cdot \mathbf{r}_j \, dt' \]

Low: dark colors; high: light colors

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

\[
\Delta E_{oa} = -\int_0^T E(t) \cdot \sum_{j=1}^{ZN_a} \sum_{i=1}^{Na} r_j \Theta(|r_j - R_i| - r_{WS}) \, dt
\]

“outer absorption” $\Delta E_{oa}$
Summary

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

• Laser-cluster dynamics is extremely complex due to the interplay between inner ionization, outer ionization, cluster expansion, and coupling to the laser field. Rather sensitive dependence on the laser and cluster parameters.

• 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)
Reserve slides
Dynamical ionization ignition

TDDFT
1D model, 9 ions, $Z = 4$
800 nm
$\hat{E} = 0.062 \ (1.3 \times 10^{14} \text{ W/cm}^2)$
$(3, 8, 3)$-pulse

Polarization enhances further inner ionization and absorption.

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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<tbody>
<tr>
<td>real $\mathcal{E}_I$</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>
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<td>model $\mathcal{E}_I$</td>
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<td>0.55</td>
<td>1.11</td>
<td>1.63</td>
<td>2.22</td>
<td>2.65</td>
</tr>
</tbody>
</table>

Fields inside the cluster at 100 nm

Ionization ignition after thermionic emission of electrons
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