Transient stability and correlation in the Coulomb explosion of large systems

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PhD 2014 predoc predoc

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<u>Einite</u> **Systems**

Critical stability IV, Dresden 2005

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Fast electron & ion dynamics in extended systems under short intense light pulses

Why?

- §*Novel light sources (XFELs) deliver unprecedented high spatial energy density in short pulses –*
- ■*clusters are an ideal target to probe this type of light-matter interaction*

large amount of photon energy transferred to the cluster in a short time

§*extreme non-equilibrium (dissipative and inhoherent!) dynamics*

- §*classical approach possible (augmented by quantum photo and Auger rates)*
- §*new phenomena ?*

Ultrafast, intense XUV to X-ray light: Multi-electron multi-photon absorption

Single photon absorption of *many* atoms in an extended but finite system: Very different from familiar multiphoton – few-electron processes in the near IR.

(Photo- Auger, and field ionzized) electrons:

➜ *Nano plasma formation*

What happens when an intense X-ray light pulse hits the cluster ?

- **Energy absorption from light leads to loss of electrons**
	- \rightarrow ionic charge builds up
	- → bound electrons from surface atoms are field ionized; electrons are trapped and form a (quasi-neutral) plasma → non-screened surface ions explode

How do we compute the dynamics ?

- § classical propagation of all charged particles (FMM if necessary)
- random photo- and Auger events (in accordance with quantum atomic or molecular rates)
- one active bound valence electron per ion, soft core Coulomb potential

Overview

Many photons quickly delivererd (X-ray):

- ◆ Massively parallel ionization (electrons): *correlation in the continuum*
- ▶ Granularity peak in ionic Coulomb explosion (ions) *most shock waves are not shock waves*…
- ▶ transient stabilization of molecular backbone in Xray induced Coulomb explosion of hydride clusters $(H₂O, NH₃, CH₄,...)$ through proton ejection *good for single molecule X-ray imaging*

Fig. 2b the PES is plotted in terms of the Scale Scale in terms of the Scale Massively parallel ionization: Sudden ionization of 100 electrons… ²*N/R* ⇥ of all activated electrons potential. In Elidden io Sudden formzation of the electrons photo active activated with excess energy **Photo** active *r*⇤ from the center is *V*mf(*r*⇤) = *V* (*r*⇤) + *V*ee(*r*⇤), where *V* level with an excess energy of *E*? = 0*.*4 keV. b) Coulomb com-

energy by the depth of the ionic background potential

mean-field result (dashed lines in Fig. 2) which can be

 \mathbf{I} at \mathbf{I} at half maximum maximum

of the charge to extension ratio of the Coulomb complex. FIG. 3: Color online: (a) Final electron spectra for *N* = 10² Vertical lines indicate excess energy ⇤ = 2. Ch. Gnodtke etal., Chem. Phys. online (2012) of the mean-field spectrum is given by the mean-field spectrum is σ in the similar to its count-field spectrum is σ

0.2 0.3 0.4 energy *E* [keV]

massively parallel ionization. a) Ar¹⁴⁷ with icosahedral geometry. Two thirds of the atoms loose an electron from the 3p

0

Massively parallel ionization correlation in the continuum

- numerical

Massively parallel ionization as a sum over binary collisions

For each electron i we sum over "virtual" binary collision contributions with all partners j ■ Can be done *analytically* using the conserved angular momentum and Runge Lenz vectors

Realize massively parallel ionization *experimentally with an attosecond pulse ?*

2nm H₂ cluster (~500 molecules), 2.5 x 10¹⁶Wcm⁻² @ ω =75 eV, T = 0.5 fs

Granularity peak in extreme ionic Coulomb explosion

n Assume that from each atom in a cluster one electron has been removed -

continuum (plasma) theory predicts:

- if electrons are removed slowly ("long" pulse): a shock wave forms
- if removal happens fast ("short" pulse): a characteristic $E^{1/2}$ dependence of the ion spectrum results
- ? is this really true for a realistic system containing a finite number of ions ?

■ depends on sharpness of cluster edge (ion distribution) § *can be tuned by pulse length of light !*

Uniform (constant) ion density ("short" pulse: complete ionization before CE)

radius r **energy E**

density
$$
\rho(r) = \frac{3}{4\pi R^3} \Theta(R - r)
$$

\nforce $f(r) = \frac{4\pi}{r^2} \int_0^r dr' r'^2 \rho(r')$
\n $= \frac{r}{R^3}$

$$
\frac{dP}{dE} = \Theta(E^*-E) \frac{3}{2} \sqrt{E/E^{*3}}
$$

Homogeneous non-uniformly decreasing ion density ("long" pulse: CE starts before ionization is finished)

density
$$
\rho(r) = \frac{1}{(\sqrt{\pi}R)^3} e^{-(r/R)^2}
$$

\nforce $f(r) = \frac{4\pi}{r^2} \int_0^r dr' r'^2 \rho(r')$
\n $= \frac{erf(r)}{r^2} - \frac{2}{\sqrt{\pi}rR} e^{-(r/R)^2}$

Kaplan etal PRL 91, 143401(03)

What *really happens* if a cluster of N ions suddenly explodes… The Contract of the 0 $\frac{1}{2}$

the pair-correlation function $\mathcal{L}_\mathcal{A}$ is shown function goal $\mathcal{L}_\mathcal{A}$ is shown function $\mathcal{L}_\mathcal{A}$

2

abundance *dP*/*dE*

(c) *N*=10 000

tion. A reduced force is expected at the surface \sim 19 \sim 2 pair-correlation function *g(r)* mean field ______ mean field with correlation hole a correlation hole in the maximum ion energy in the mean-field \sim maximum ion energy using Eq. (8) in the maximum ion energy in the maximum io \mathbf{r} in can neighborrow show as solid line (b). Additionally, \mathbf{r}

- 1000 atoms in an LJ cluster $\mathsf{B}^\mathsf{r}_\mathsf{S}$ is the shell s
- scales from comparable shells with routinuum system CS: with a reas (a)–(c). The mean-field areas (a)–(c). The mean-field areas (a)–(
Shells with Gauss' law for gray-shaded areas (a)–(c). The mean-field areas (a)–(c). The mean-field areas (a)–($\mathbf e$ scales from comparison with N % 1000 in the N
- \bullet CS doubles size in τ is shown with its shown with dashed lines. CS doubles size in τ
	- e maximum ion energy E^{*}

particle at r2 in Fig. 4). Both expectations are confirmed by the confirmed by the confirmed by the confirmed b
The confirmed by the conf

this shell. Yet, this modification of the forces does not

play a role as long as the test particle's correlation hole

What *really happens* if a cluster of N ions suddenly explodes… The Contract of the Physical Review Letters week end in the contract of the contract

(c) *N*=10 000

2 pair-correlation function *g(r)* mean field mean field with correlation hole

looks like a shock wave, but should not:
but should not: the sketch in Fig. 4 and its inset, which shows which shows in Fig. 1. And its inset, which shows in the shows $\frac{1}{2}$ ing the integration over $\frac{1}{2}$ yields—as expected—Gauss' $\frac{1}{2}$

homogeneously charged sphere just replaced by (true) discrete the seed in Fig. 3. \blacksquare ions of the same density **inside the shell with radius relations** $\sum_{i=1}^n\frac{1}{i!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{1}{j!}\sum_{j=1}^n\frac{$

• 1000 atoms in an LJ cluster

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- scales from comparable continuum system CS: with row in accordance with \sim r in accordance with \sim r in accordance with \sim
- \bullet CS doubles size in τ
- e maximum ion energy E^{*}

Force from a spherical shell of charged particles (at $r = 1$)

Time dependence distinguishes shock wave from granularity peak

Crossover from *granularity* to *shock wave* dominated dynamics PRESENTENT 110, 133401 (2013) PHYSICAL REVIEW LETTER WEEK ENDING: A PHYSICAL REVIEW LETTERS were also assumed to the contract of the contract

decided by a/δ in the charge reaching a maximal value at \sim rmax ¼ R \$!=3 (for ! ' R).

similar expansion, i.e., scaling of all lengths in the system

with the common factor \mathcal{C} . Consequently, the force (7) is the

ratio of softness of cluster edge a (modeled by Fermi distribution) late to versus correlation hole size δ $\frac{1}{\sqrt{1-\frac{1}{n}}}$ in the of the properties of the property of the property of the property of the property of th final ion-energy spectrum, we may even attention to calcuand a (modeled by Fermi distribution) $\frac{1}{2}$ with isolation $\frac{1}{2}$ is given in unitary in the correlation hole radius $\frac{1}{2}$

of the correlation hole radius \mathcal{C} the correlation hole can see the crossover \mathcal{C}

 \mathcal{F}_1 (color online). Energy spectra as obtained from an obtaine exploding cluster with 1000 particles for various cluster with 1000 particles for various cluster edges in the

Ion bunching through granularity

- dynamics of exploding ionic nanoplasmas quite rich due to the combination of finiteness with granularity
- difficult to disentangle with "final" energy spectra:

 \rightarrow probe short time dynamics of expanding ionic plasmas

U Saalmann, A Mikaberidze and JM Rost, PRL 110, 133401 (2013)

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Methane clusters (N~1000) under 1keV, 10fs X-ray pulses

Electron and ion dynamics in time namely, the ratio of the average energy of all protons versus $\mathsf{CS}\ \mathsf{In}\ \mathsf{time}$

indicates a dynamical segregation of protons and heavy ions as can be seen in Fig. 3, where a more global quantity,

Electron and ion dynamics in time namely, the ratio of the average energy of all protons versus $\mathsf{CS}\ \mathsf{In}\ \mathsf{time}$

H2O

ic proton ojection co **• why is proton ejection so** Fig. 2 (colores to intensity ? The fastest intensity \mathcal{P} 0.5 ps after the peak of the pulse (T α), versus the average (T α), versus the average α

number of photons absorbed per atom/molecule n.m. \mathcal{L}^{max} Cluster size is N ¼ 689. The kinetic energies are normalized

Kinetic energy

indicates a dynamical segregation of protons and heavy ions as can be seen in Fig. 3, where a more global quantity,

Protons and heavy atoms according to Eq. (3) for X \sim **0) for X** \sim **0)** \sim **0)** \sim **0)** \sim $\frac{1}{2}$ function of the x-ray international parameters as in Fig. 2. Same parameters as in Fig. 2. gation is a consequence of the consequence of the cluster nature of the cluster nature of the cluster nature of dentite system. To this end, we have set up a simple model we have set up a simple model we have set up a simple mod

- ions don't move
- **Extrapped electrons stay cool** \blacksquare trapped electrons stay cool \blacksquare

 \mathcal{F}_1 (color online). Ratio K online). Ratio K online). Ratio K of the average kinetic energy of the average kinetic ener

→ *protons must carry away* **the energy !? higher mass of the mass of the energy !? For protons must carry away** $\frac{1}{\sqrt{2}}$ is the change in $\frac{1}{\sqrt{2}}$ in $\frac{1}{\sqrt{2}}$ in $\frac{1}{\sqrt{2}}$ in $\frac{1}{\sqrt{2}}$

that of all heavy ions (X α α β β

$\langle \chi_{\rm in} \rangle_X$ Charging/Intensity dependence: Minimal Coulomb explosion dynamics face. The mean kinetic energy is larger in Γ not take place in either of the two regions. In region B,

protons and heavy ions originate from all initial positions

in the cluster with an increasing energy towards the sur-

- of radius P propagated for 1 ps \mathbf{u} \overline{a} Abundance *P* [%] \sim radius κ , propagated for μ ps sphere of radius R, propagated for 1 ps
- 0.0125 mass m $\frac{1}{4}$ m = 20 $74.$ III – 2 **a** $\frac{3}{4}$: proton mass m_p, $\frac{1}{4}$: m = 20 x m_p
- FIG. 5 (color online). Charge-state distribution for carbon ions $\overline{}$ core are light enough (or more precise: have are precise: have are precise: $\overline{}$
- r_s : screening radius x-ray intensities I and C₆89 for various x-ray intensities I and C₆89 for various x-ray intensities I and C₆89 for various x-ray intensities intensities intensities intensities intensities \mathbf{S} sufficiently large-to-mass ratio \mathbf{S}
- α to the prison is a result, the surplus of segregation is a result, the surplus of screening electrons prevents heavy ions in the surface layer even beyond the

 $\mathcal{O}(\mathcal{O})$. This is independent the case as can be seen in Fig. 5. For low low low low low low low low low

- intensities most carbon atoms remain neutral in the prior of the prior of the prior of the prior of the prior o α well as in the hydride cluster. This changes drastically constructed as α In contrast, protons escape with high final energy from the
- for intermediate intensities of about 10¹⁸ W=cm2, where → before field ionized electrons move inwards, protons *(also from the interior)* **Fig. heave the cluster, on the other hand, about 80% neutral heavy** atoms result from recombination with the cold electrons $\mathcal{L}_{\mathcal{A}}$ $\frac{32}{3}$ $\frac{1}{2}$ $\frac{1}{2}$ $\frac{1}{2}$ $\frac{1}{2}$ leave the cluster
	- after proton segregation in the surface layer, which has \rightarrow surplus of screening electrons inhibits motion of heavy ions beyond $r_{\rm s}$ and the proton segment \sim cease (see Fig. 2) and, as a consequence, single consequence, similar charges α

Proton segregation in X-ray illuminated protonated clusters

- **dynamical effect through intricate interplay of electron** screening by field ionization and early proton escape
- § *signature: hardly any charged heavy ions*
- **P** requires the right amount of charging (laser intensity)
- leads to efficient energy transport by protons (reduced radiation damage):
	- **→** heavy ion molecular backbone stays intact
	- \rightarrow nanoplasma is cold

→ good conditions for CDI coherent diffraction imaging...

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