Transient stability and correlation in the Coulomb explosion of large systems

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Pierfrancesco Di Cintio PhD 2014
Abraham Camacho Garibay predoc
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Christian Gnodtke (now City Bank London)
Alexey Mikaberidze (now ETH, Zurich)
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Critical stability IV, Dresden 2005

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mpipks

Finite Systems

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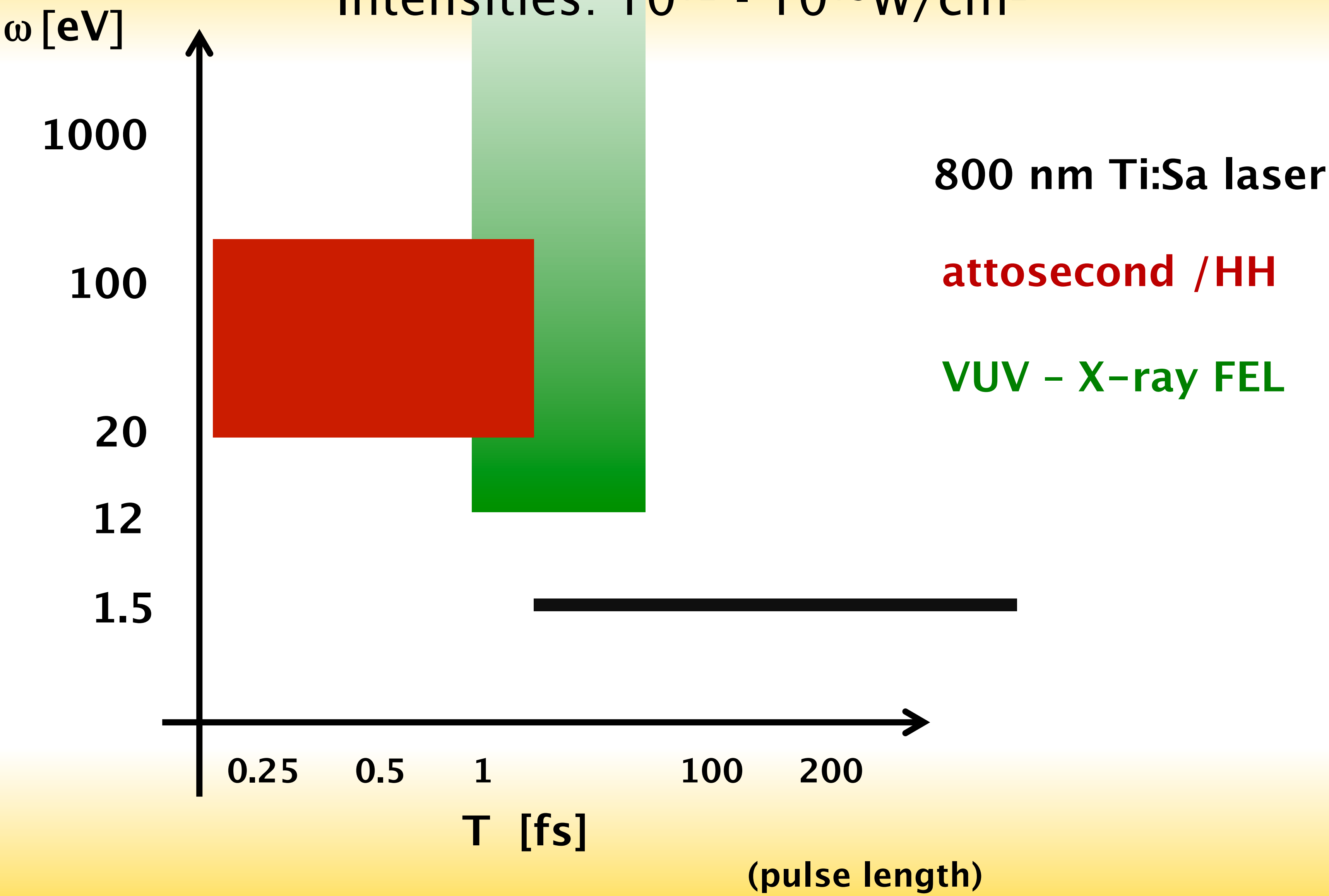
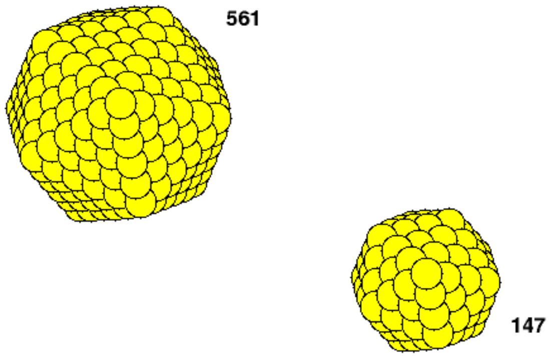
Light pulse parameter space
Intensities: 10^{12} - 10^{19}W/cm^2



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

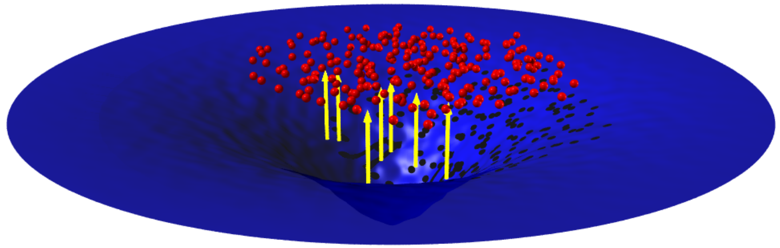
- *extreme non-equilibrium* ***(dissipative and inhoherent!)*** *dynamics*
- *classical approach possible (augmented by quantum photo- and Auger rates)*
- *new phenomena ?*

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 ?

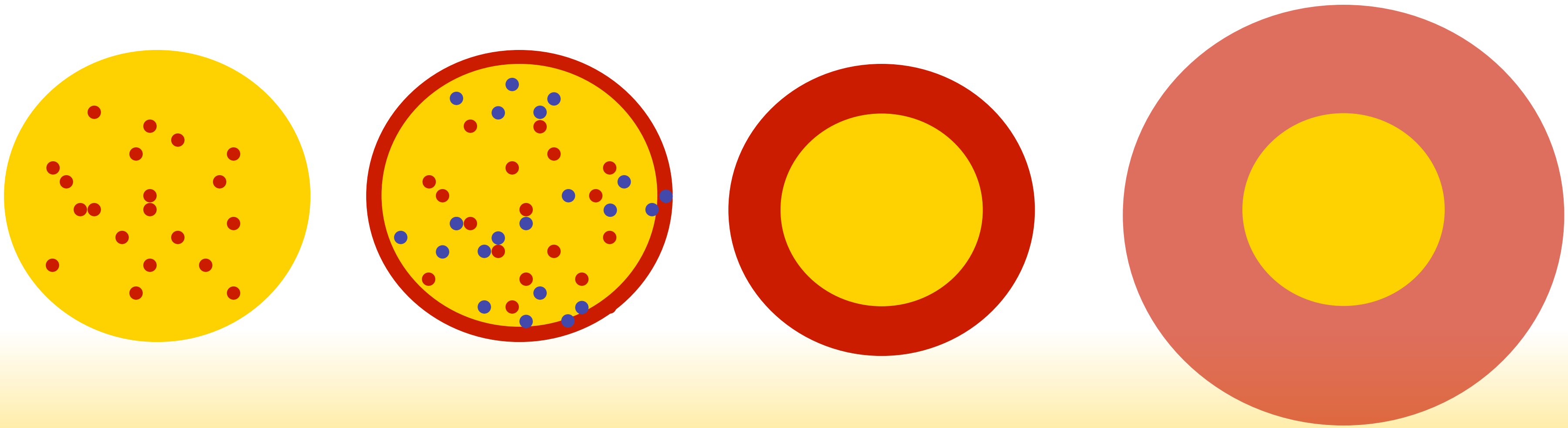
- Energy absorption from light leads to loss of electrons

➜ ionic charge builds up

➜ bound electrons from surface atoms are field ionized;

electrons are trapped and form a (quasi-neutral) plasma

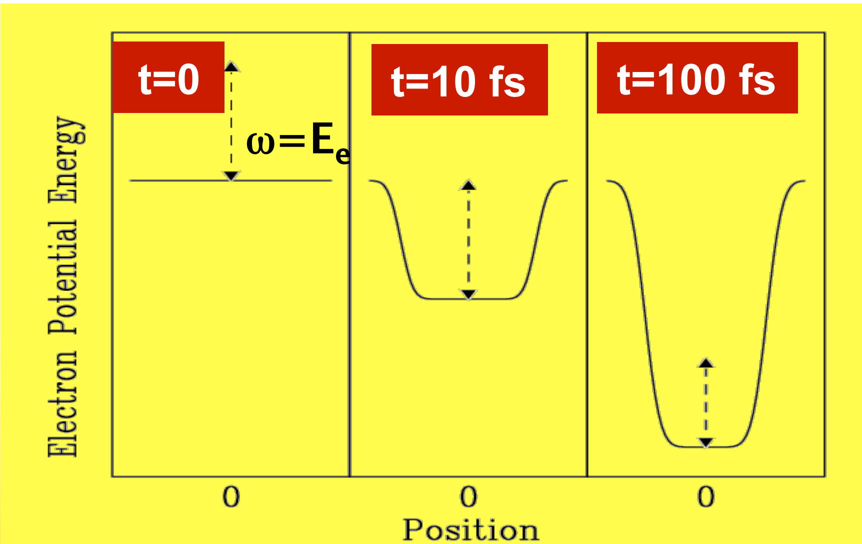
➜ non-screened surface ions explode

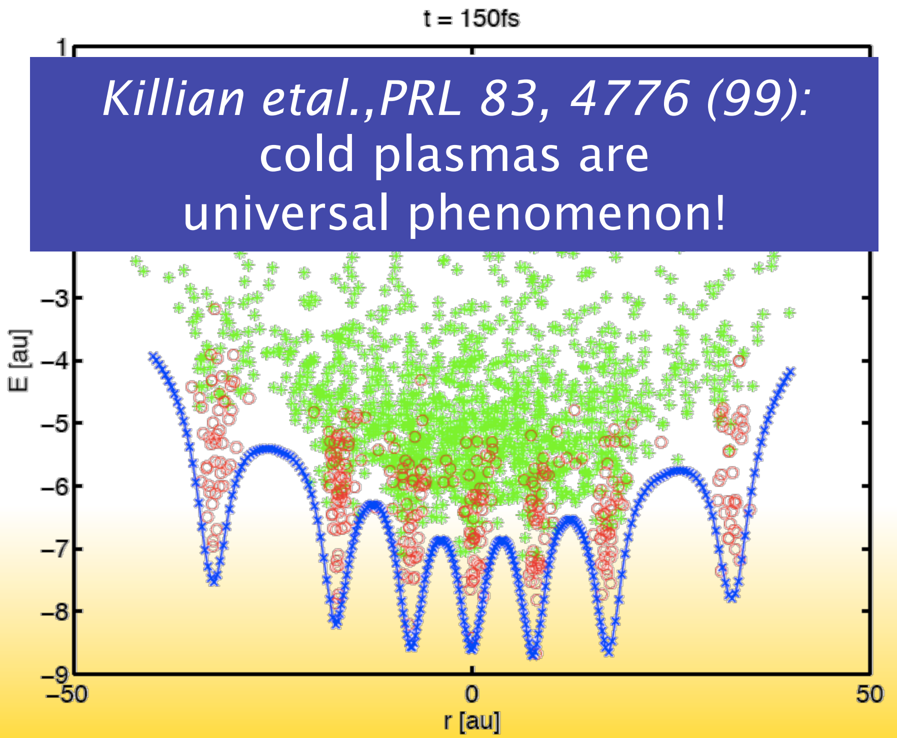


How do we compute the dynamics ?

- 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

t=0 **t=1ns** **t=1μs**





Killian etal.,PRL 83, 4776 (99):
cold plasmas are
universal phenomenon!

Overview

Many photons quickly delivererd (X-ray):

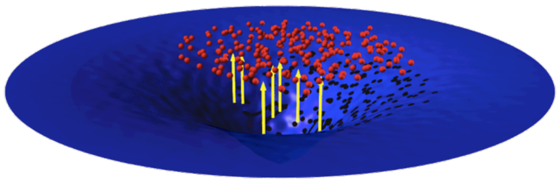
- Massively parallel ionization (electrons):
  correlation in the continuum

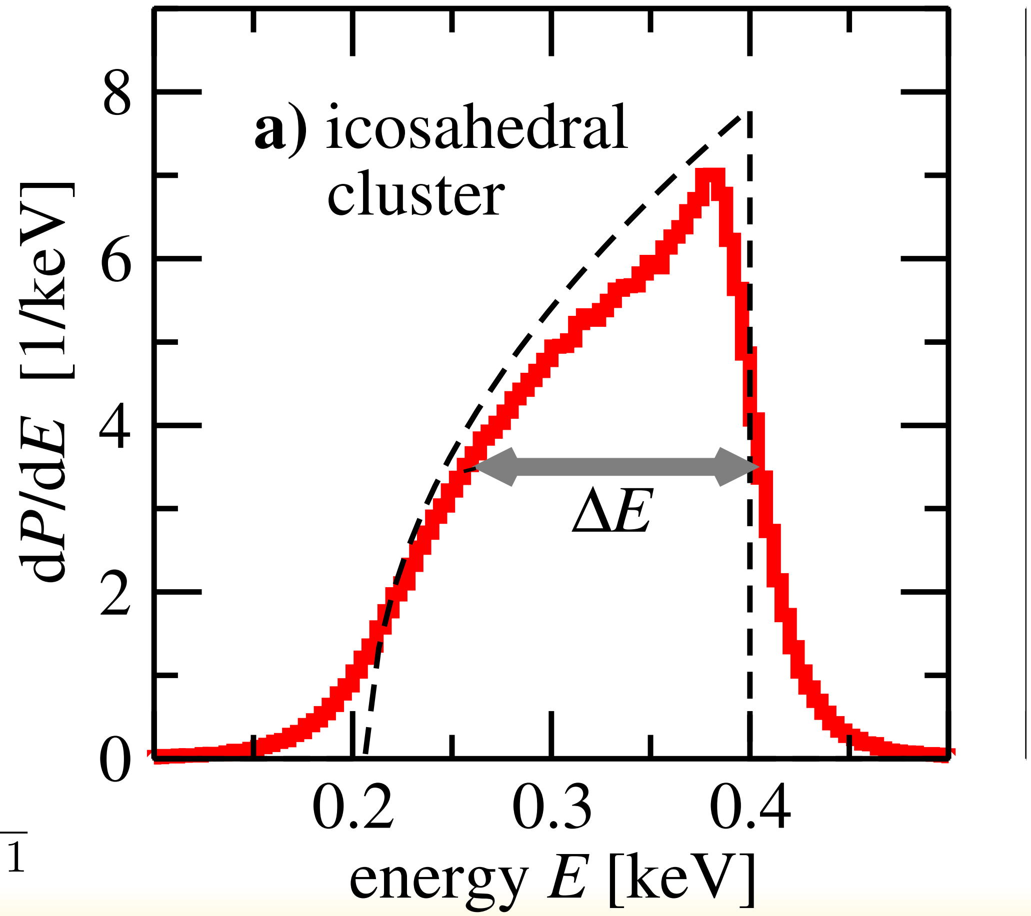
- Granularity peak in ionic Coulomb explosion (ions)
  most shock waves are not shock waves...

- transient stabilization of molecular backbone in X-ray induced Coulomb explosion of hydride clusters (H_2O, NH_3, CH_4,...) through proton ejection
  good for single molecule X-ray imaging

Massively parallel ionization:
Sudden ionization of 100 electrons…

Ar_{147}, 100 e^- suddenly activated with 400 eV excess energy



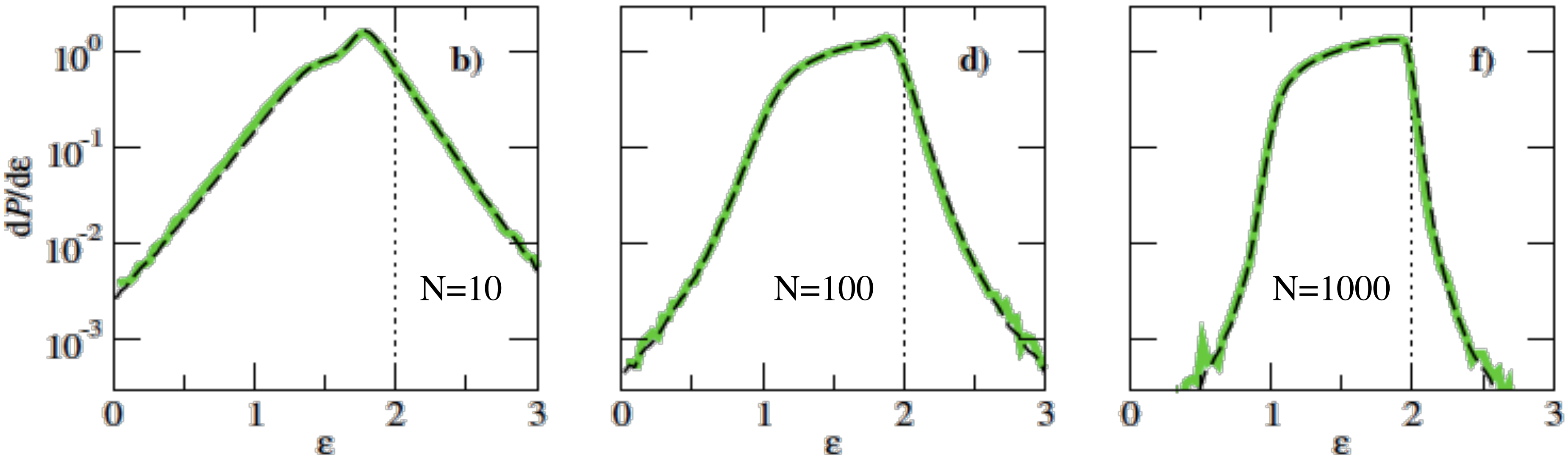


- Mean field: $$\frac{dP(\varepsilon)}{d\varepsilon} = \frac{3}{2}\sqrt{\varepsilon - \varepsilon^* + 1}$$

- FWHM: $$\Delta E_{\mathrm{FWHM}} = \frac{3}{4}V_0 = \frac{9}{8}N/R$$

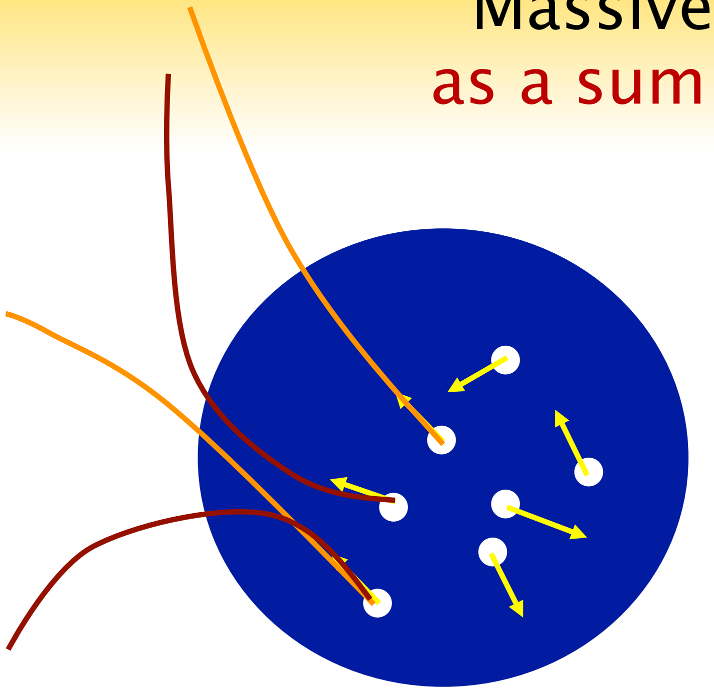
Ch. Gnodtke etal., Chem. Phys. online (2012)

Massively parallel ionization
correlation in the continuum

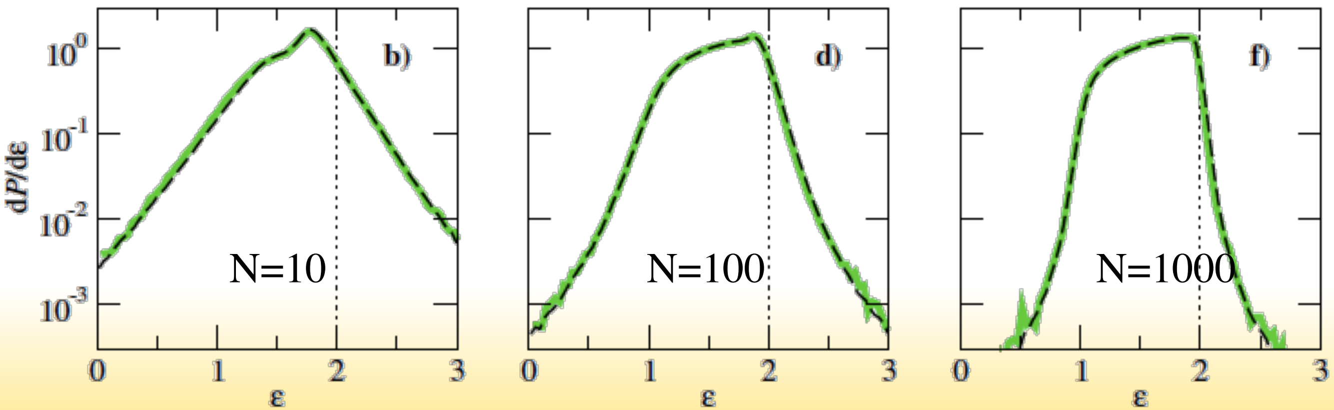


--- analytical,
-   numerical

Massively parallel ionization
as a sum over binary collisions



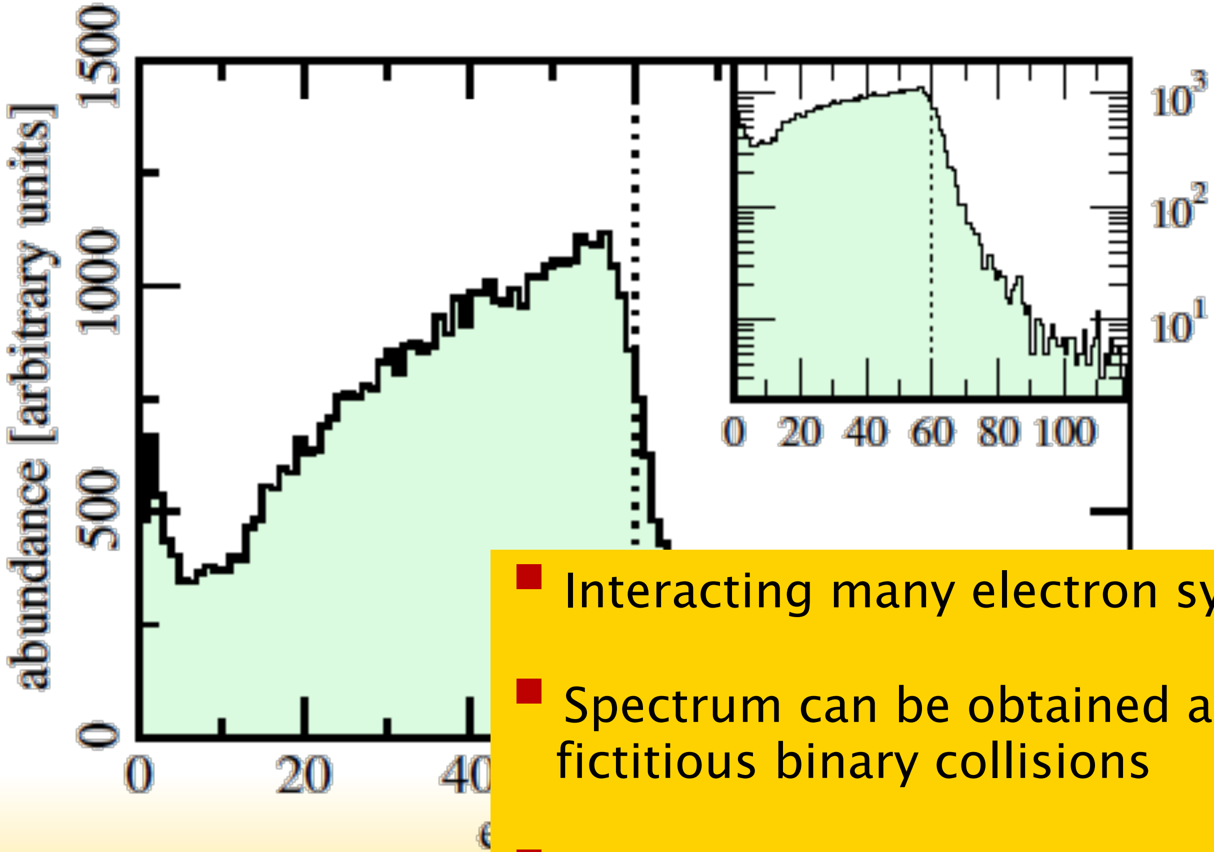
- For each electron i we sum over "virtual" binary collision contributions with all partners j
- Can be done *analytically* using the conserved angular momentum and Runge Lenz vectors



--- analytical,
- numerical

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

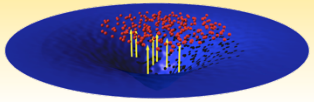
2nm H_2 cluster (~500 molecules), 2.5 x 10^{16} Wcm^{-2} @ ω=75 eV, T = 0.5 fs



- Interacting many electron system
- Spectrum can be obtained analytically from fictitious binary collisions
- generic feature under short intense X-ray pulses

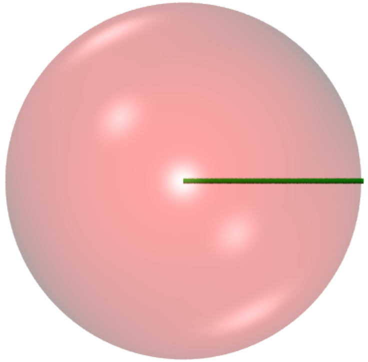
So far electron spectra…
… what about the ions ?

Granularity peak
in extreme ionic Coulomb explosion

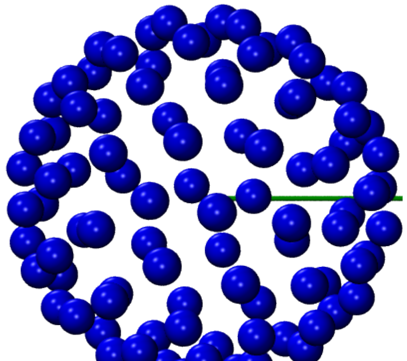


- Assume that from each atom in a cluster one electron has been removed -

  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

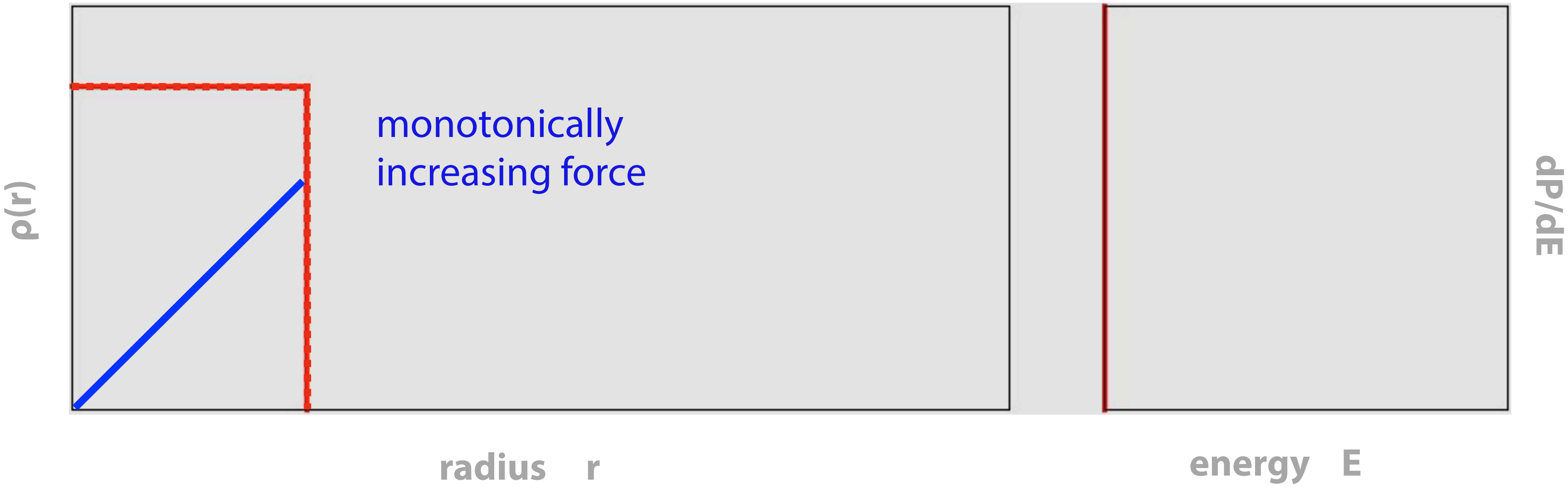


? is this really true for a realistic system containing a finite number of ions ?



- 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)

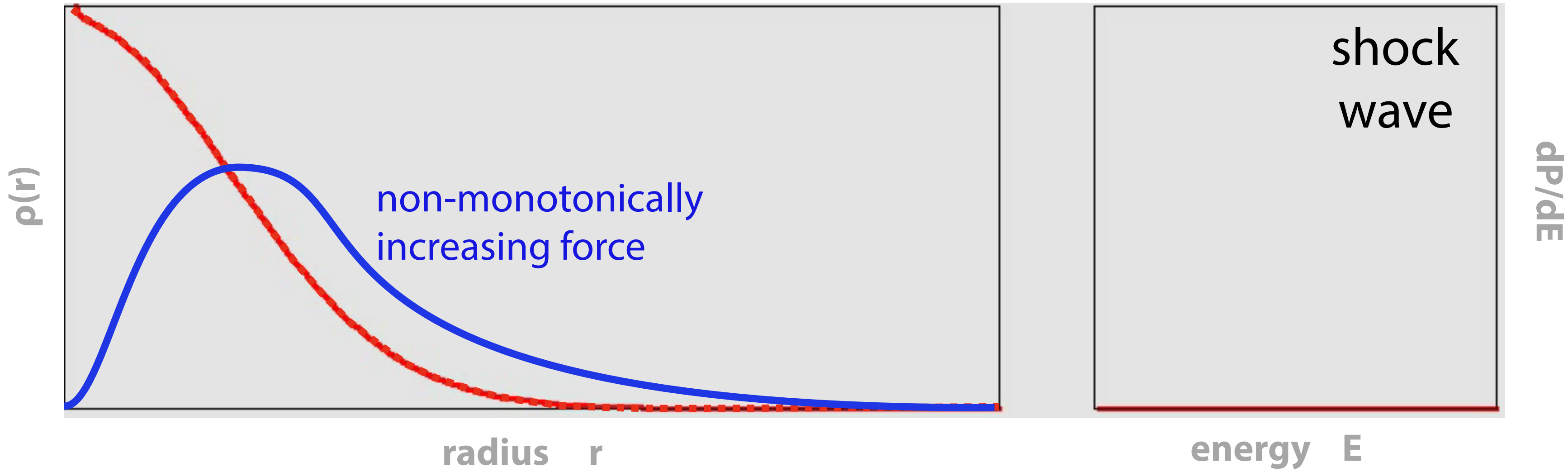


density $$\rho(r) = \frac{3}{4\pi R^3}\Theta(R-r)$$

force $$f(r) = \frac{4\pi}{r^2}\int_0^r \mathrm{d}r' r'^2 \rho(r') = \frac{r}{R^3}$$

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

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

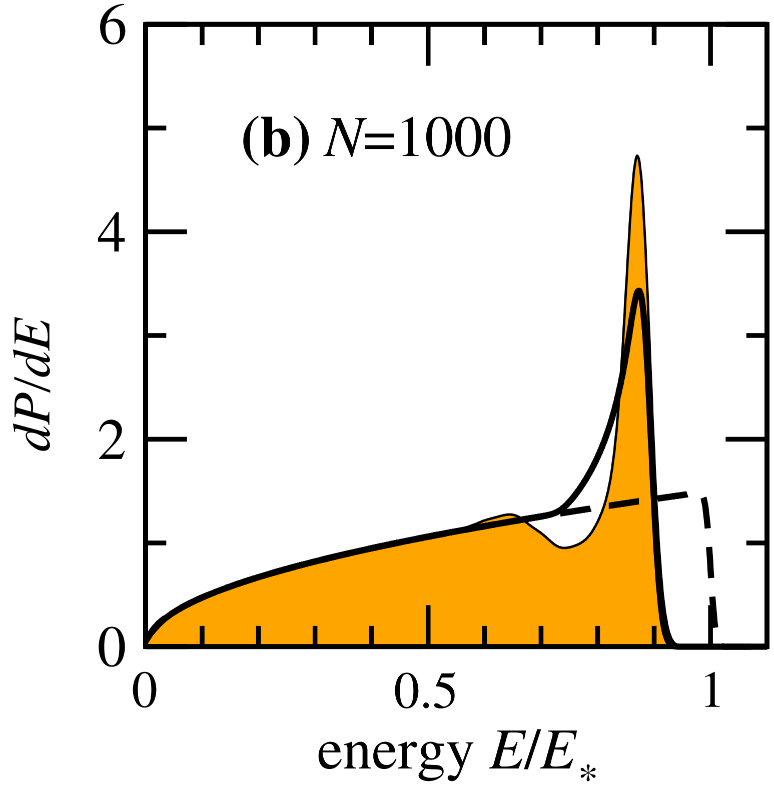


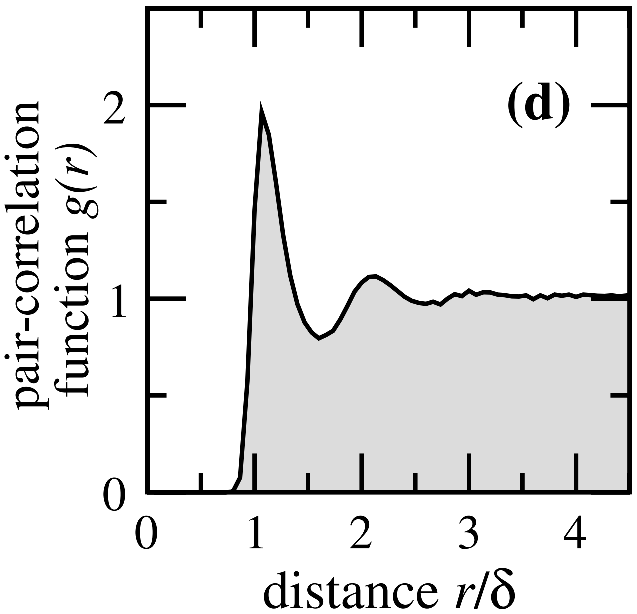
density $$\rho(r) = \frac{1}{(\sqrt{\pi}R)^3} e^{-(r/R)^2}$$

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

Kaplan etal
PRL 91, 143401(03)

What *really happens* if a cluster of N ions suddenly explodes…





- 1000 atoms in an LJ cluster
- scales from comparable continuum system CS:
- CS doubles size in τ
- maximum ion energy E^*

– – – mean field

______ mean field with correlation hole

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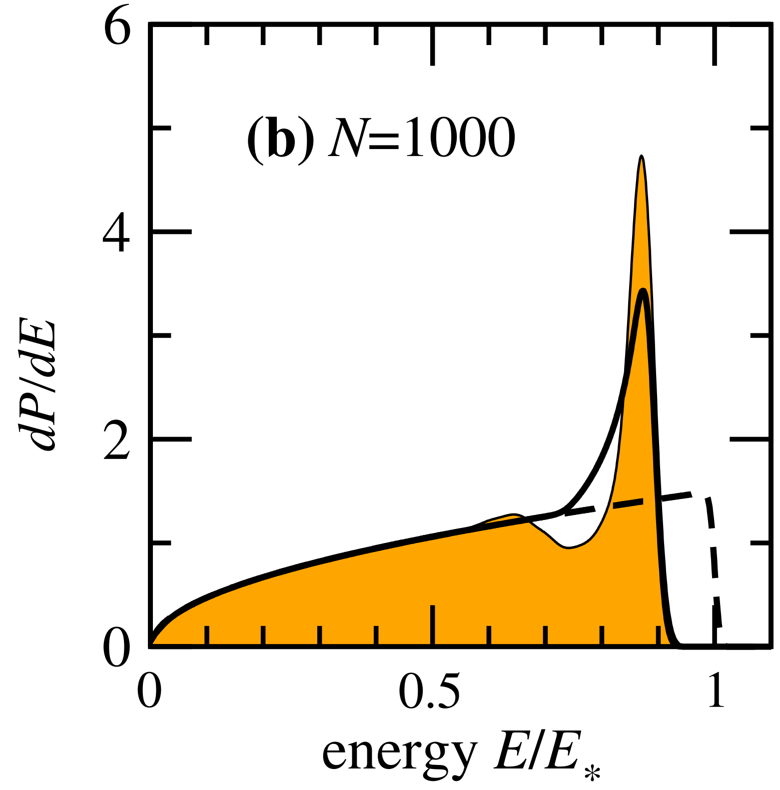

looks like a shock wave,
but should not:

homogeneously charged sphere
just replaced by (true) **discrete ions** of the same density

- 1000 atoms in an LJ cluster
- scales from comparable continuum system CS:
- CS doubles size in τ
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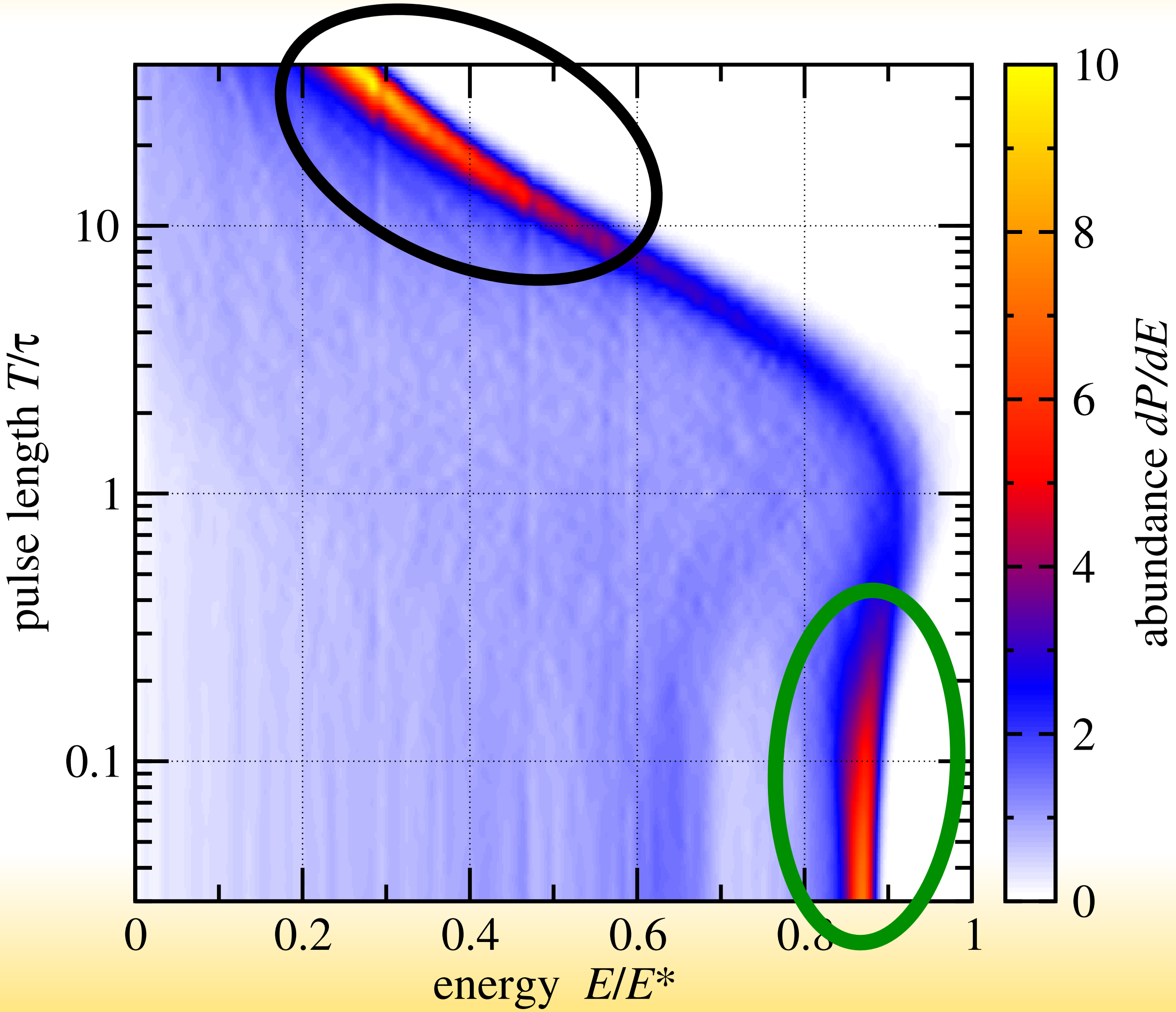
– – – mean field
______ mean field with correlation hole

True energy spectra of exploding ion nano plasmas as a function of activation time (pulse length T)



shock wave

granularity peak

- 1000 atoms in an LJ cluster
- scales from comparable continuum system CS:
- CS doubles size in τ
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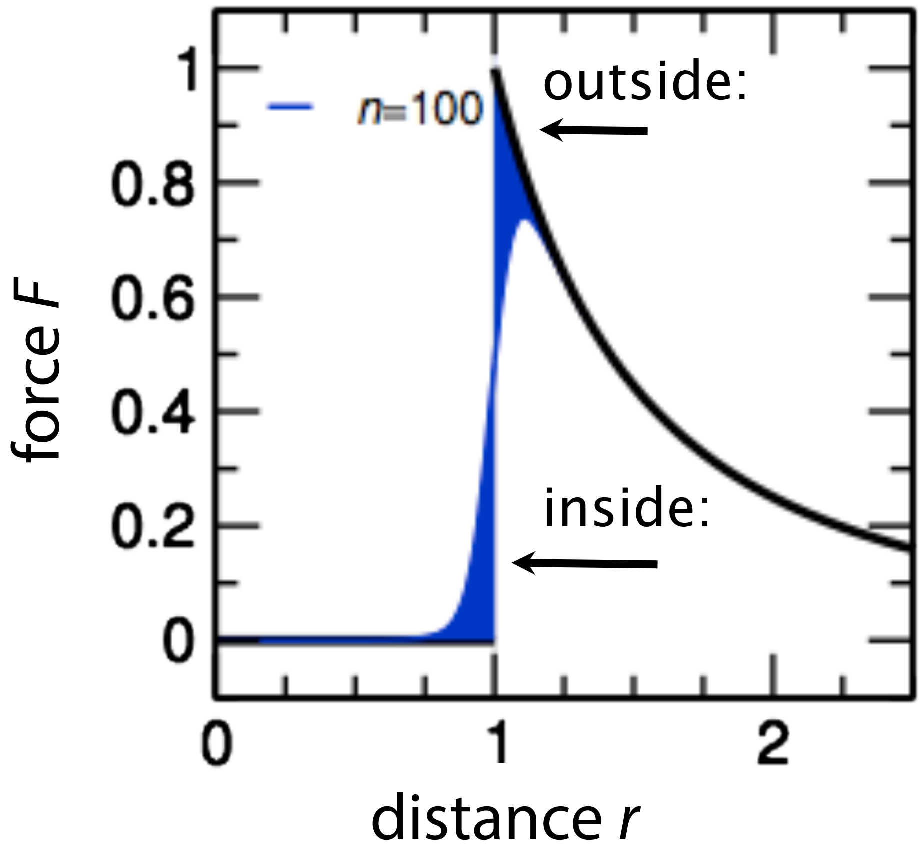
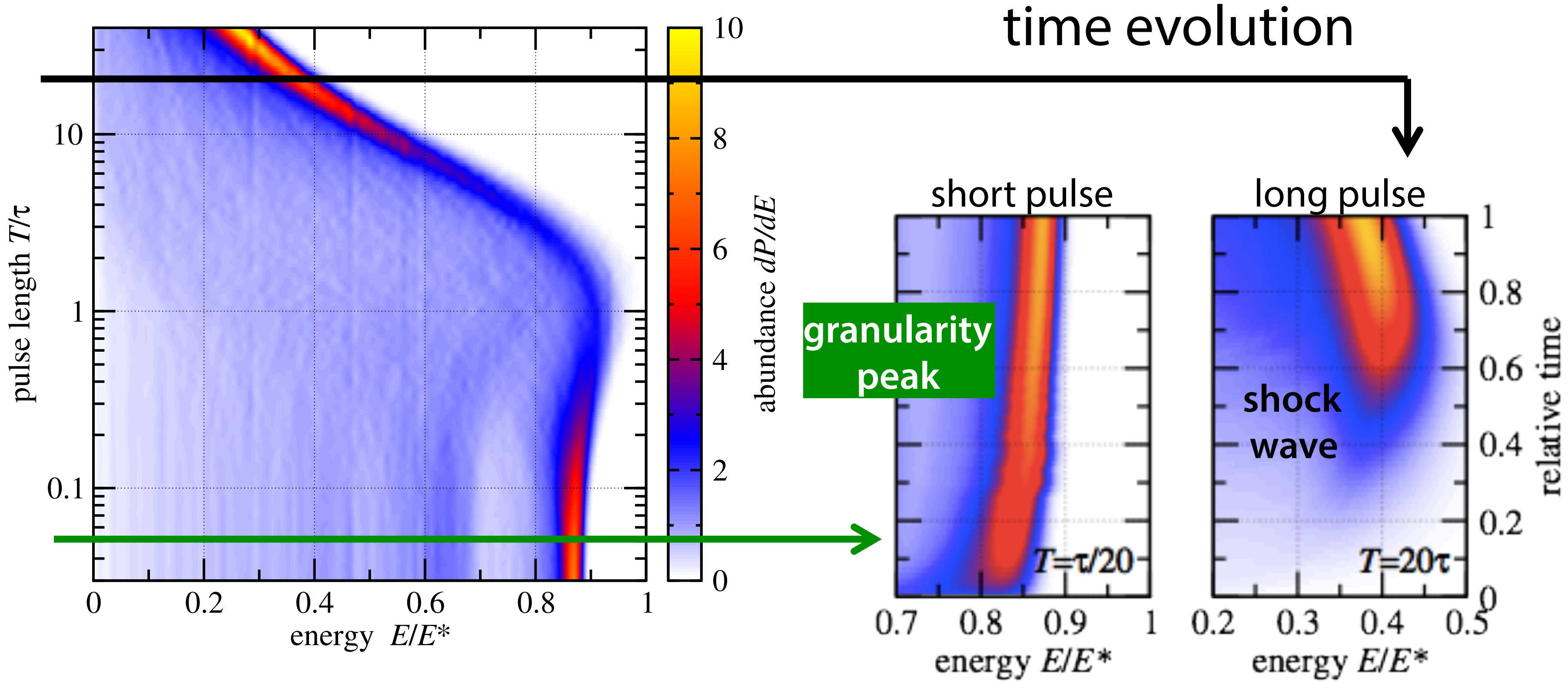
Force from a spherical shell of charged particles (at r =1)



reduced w.r.t. continuous distribution

finite force, i.e., increased w.r.t. cont. distribution

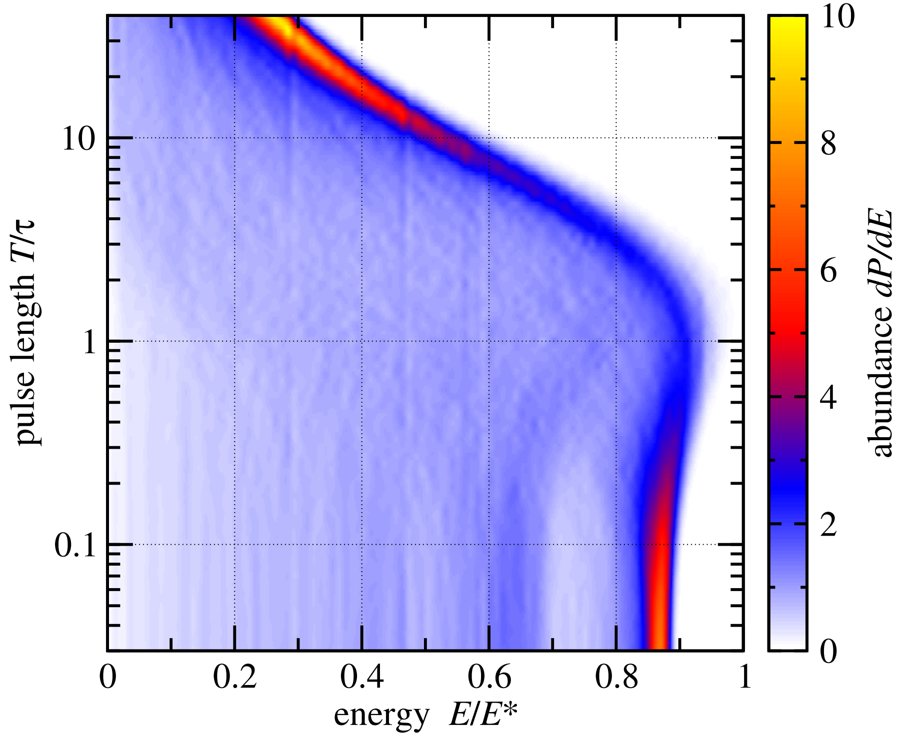
Time dependence distinguishes shock wave from granularity peak

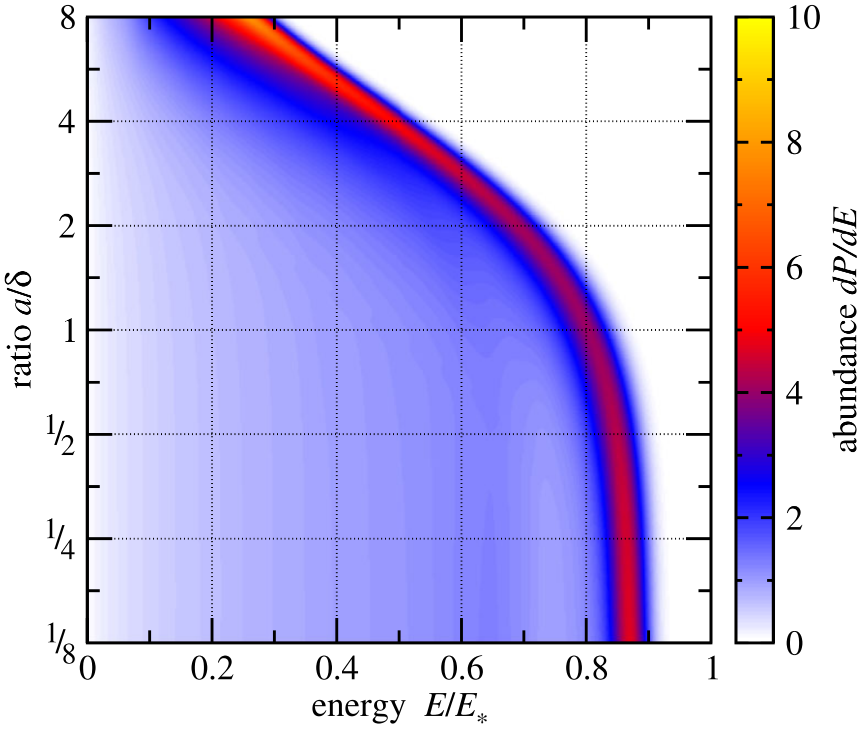


granularity peak right from the beginning

shock wave develops later

Crossover from *granularity* to *shock wave* dominated dynamics





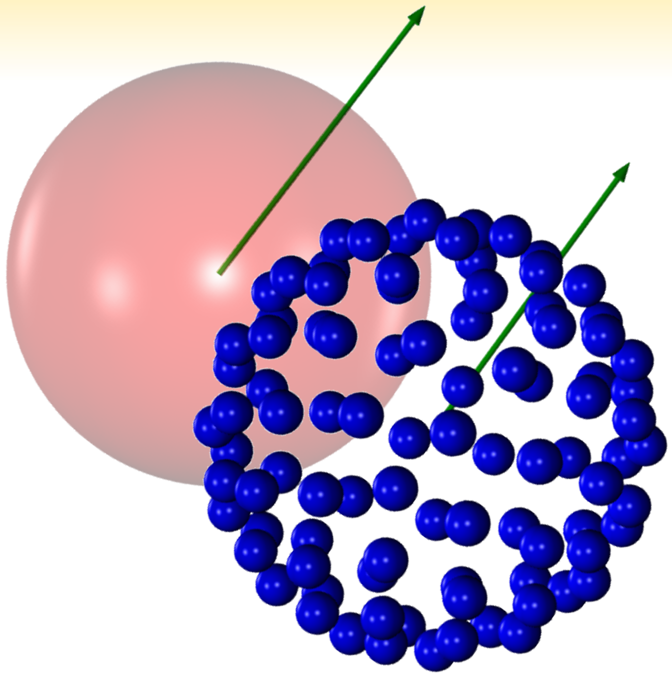
decided by a/δ

ratio of softness of cluster edge a (modeled by Fermi distribution) versus correlation hole size δ

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:



➜ probe short time dynamics of expanding ionic plasmas

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

Overview

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- Granularity peak in ionic Coulomb explosion (ions)
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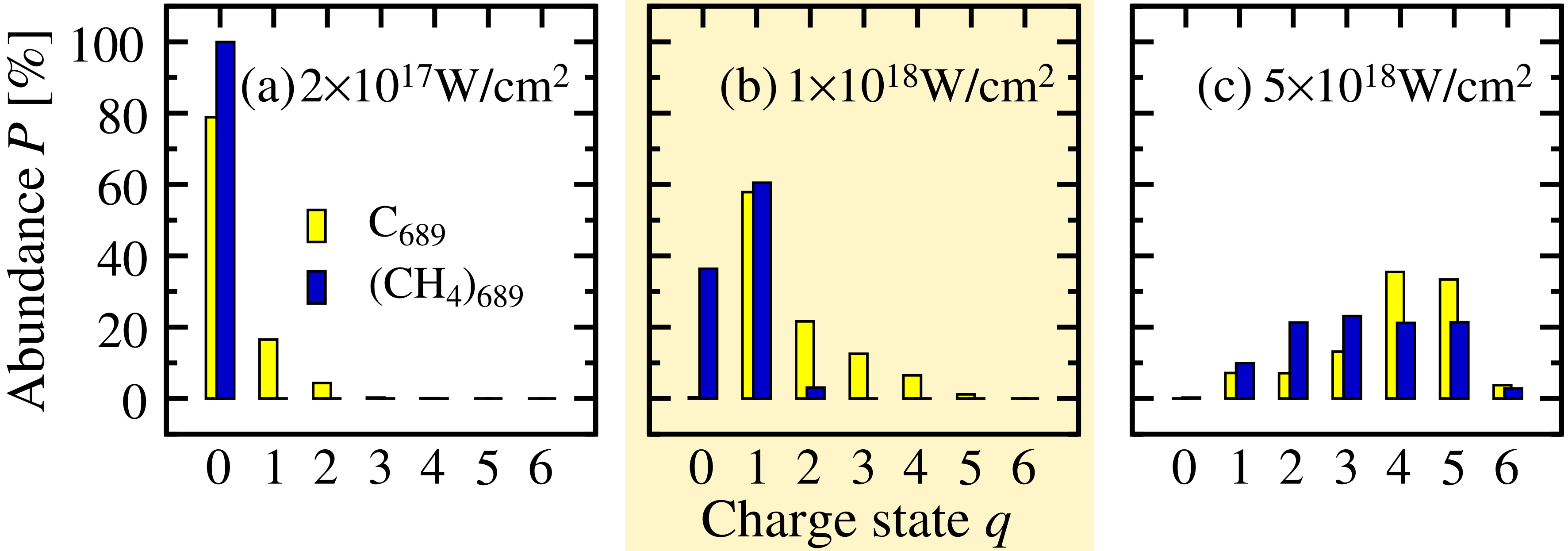
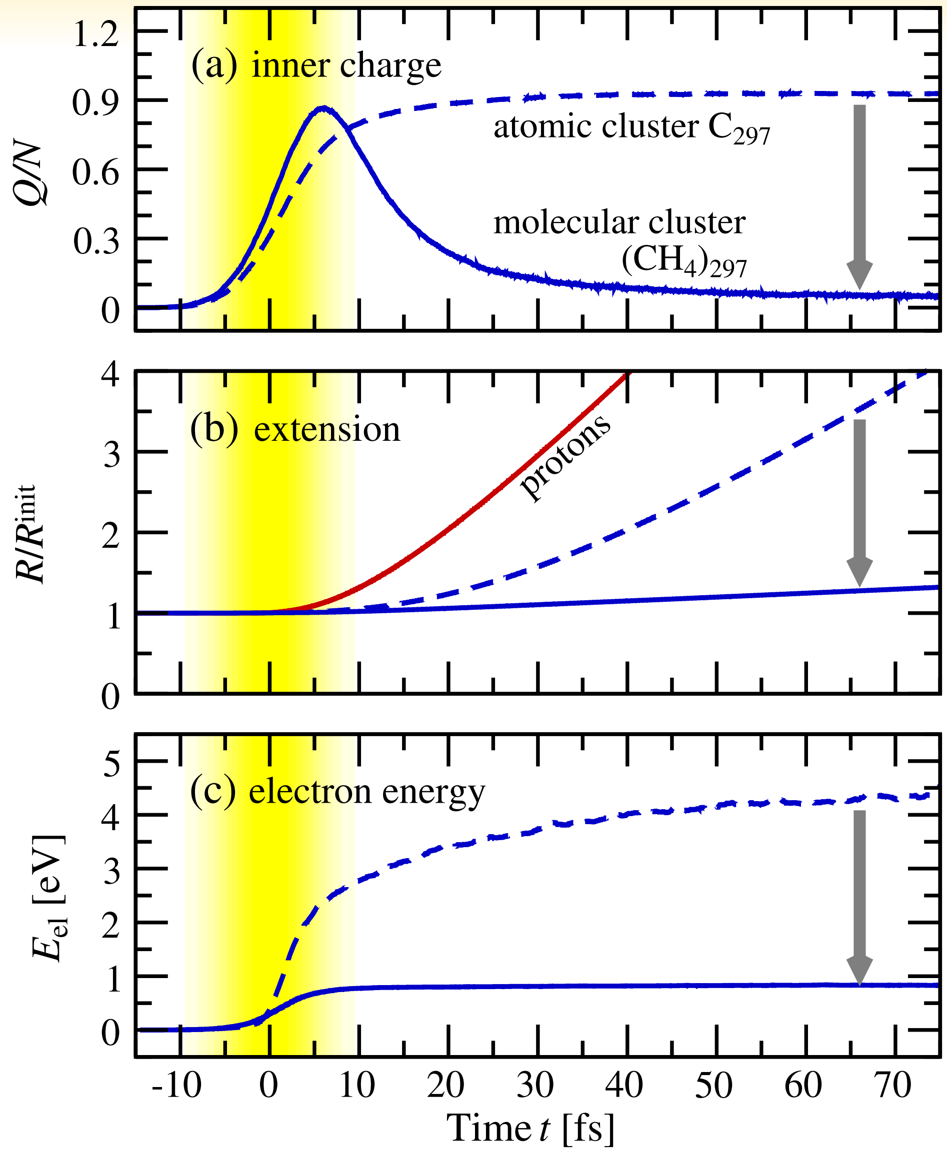
- transient stabilization of molecular backbone in X-ray induced Coulomb explosion of hydride clusters (H_2O, NH_3, CH_4,…) through proton ejection
  good for single molecule X-ray imaging

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

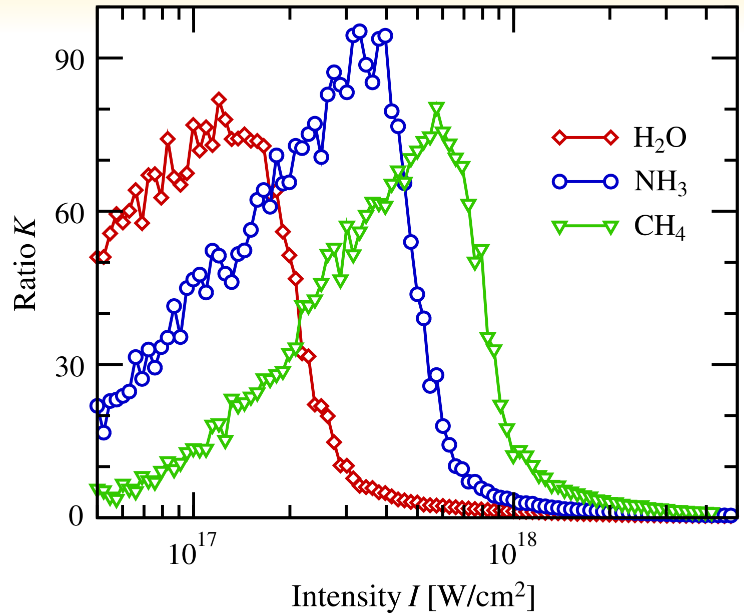


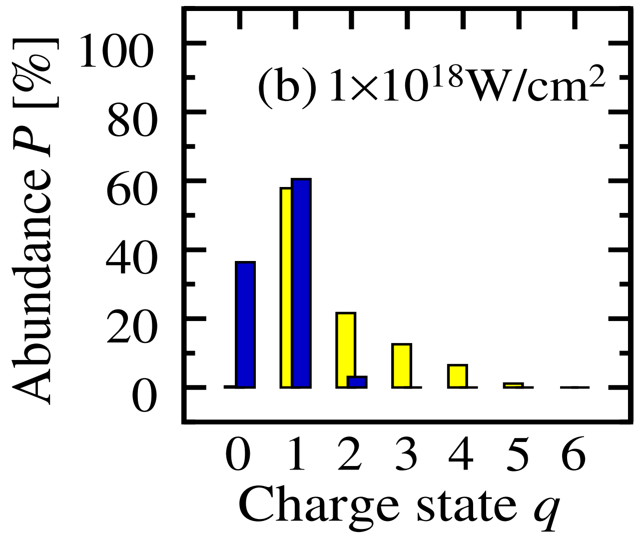
LCLS

Electron and ion dynamics in time



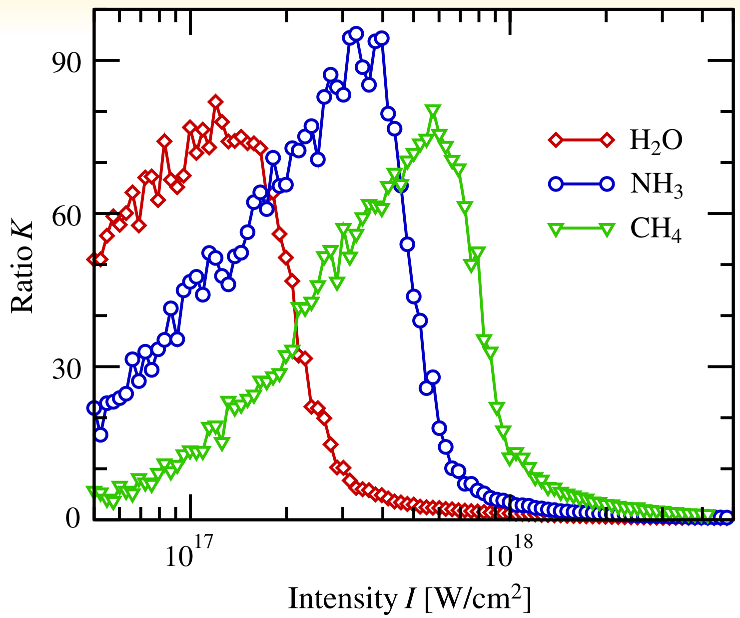
$K \equiv \langle E_{kin} \rangle_H / \langle E_{kin} \rangle_X$





Electron and ion dynamics in time

$K \equiv \langle E_{\text{kin}} \rangle_H / \langle E_{\text{kin}} \rangle_X$



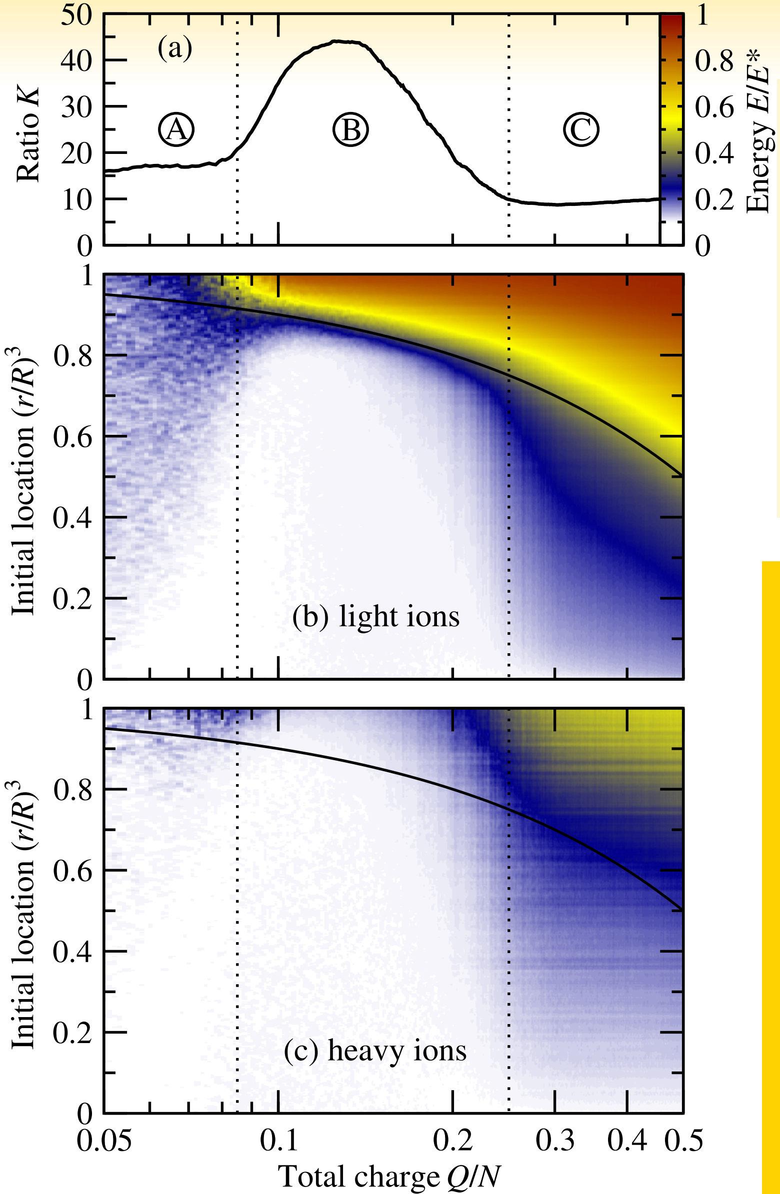
- why is proton ejection so sensitive to intensity ?

Molecular cluster:

- ions don't move
- trapped electrons stay cool

➜ *protons must carry away the energy !?*

$$K \equiv \langle E_{\text{kin}} \rangle_{\text{H}} / \langle E_{\text{kin}} \rangle_X$$



Charging/Intensity dependence: Minimal Coulomb explosion dynamics

- 10^4 ions homogeneously distributed over sphere of radius R, propagated for 1 ps
- ¾: proton mass m_p, ¼: $m = 20 \times m_p$
- N-Q electrons placed at random ions
- r_S : screening radius

- A & C: exploding ions (heavy & light) from all radii in the cluster
- B: only protons explode:

➜ before field ionized electrons move inwards, protons *(also from the interior)* leave the cluster

➜ surplus of screening electrons inhibits motion of heavy ions beyond r_S

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*
- requires the right amount of charging (laser intensity)

- leads to efficient energy transport by protons (reduced radiation damage):
  - ➜ heavy ion molecular backbone stays intact
  - ➜ nanoplasma is cold

➜ good conditions for CDI coherent diffraction imaging…

PF Di Cintio, U Saalmann & JM Rost, PRL 111, 123401 (2013) mpipks

Finite Systems

Thanks !

For pre/reprints please visit http://www.pks.mpg.de/~rost