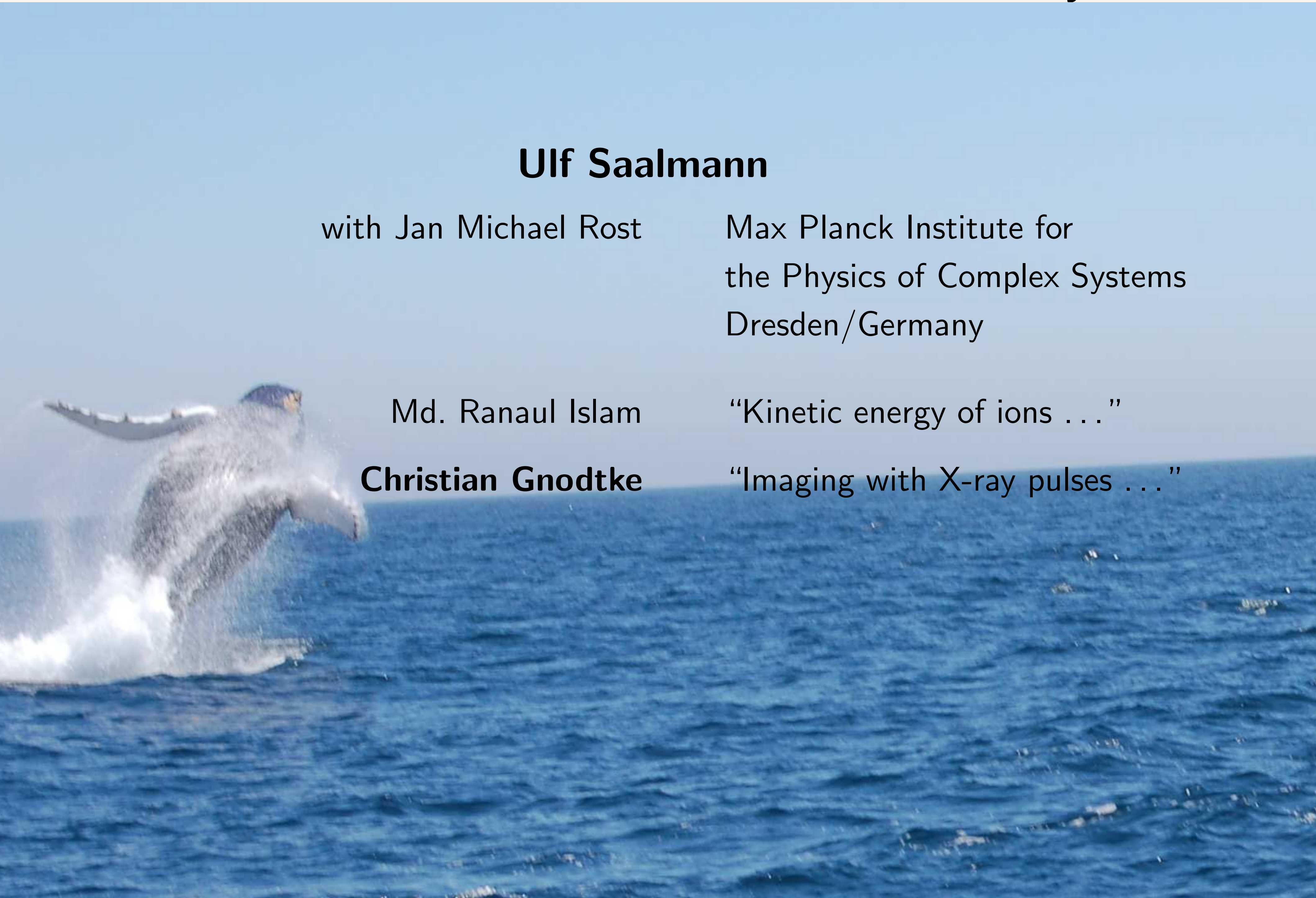
Clusters in Intense Laser Pulses: From Infrared to X-Ray Radiation



Ulf Saalmann

with Jan Michael Rost

Max Planck Institute for
the Physics of Complex Systems
Dresden/Germany

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

Christian Gnodtke — “Imaging with X-ray pulses . . . ”

outline

- 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

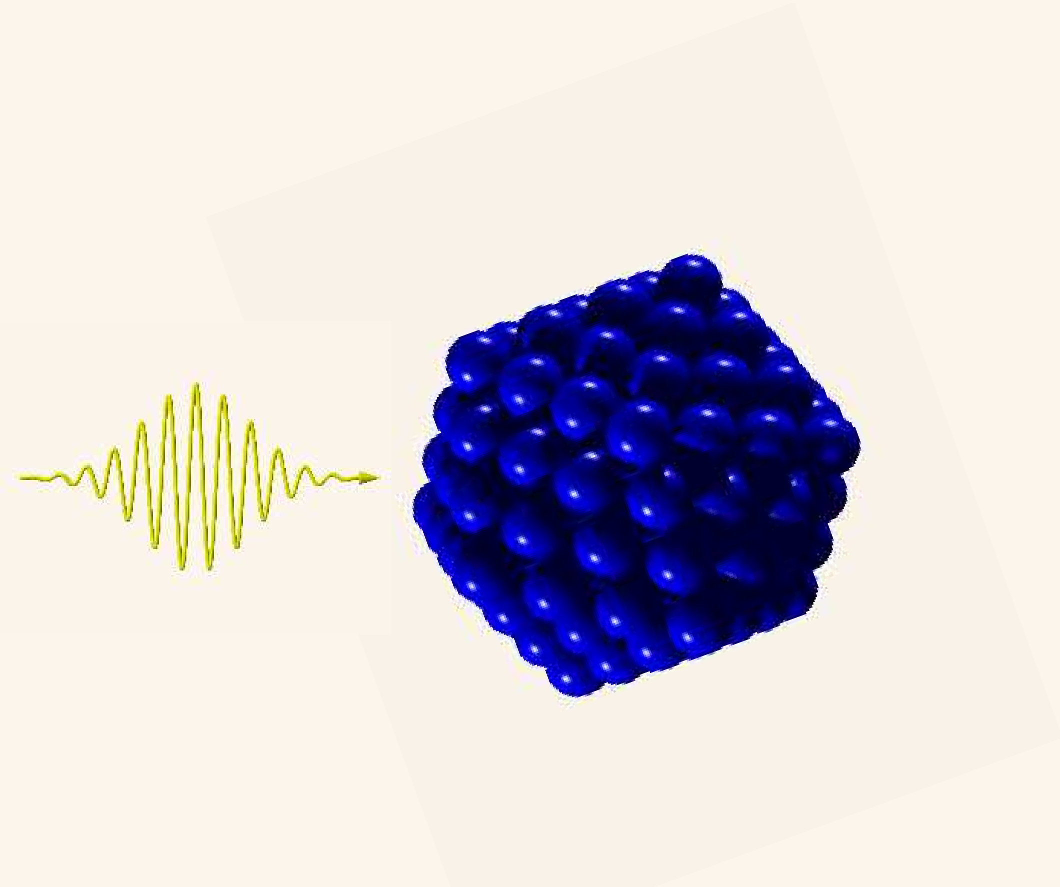
- study of properties (e. g. laser-matter interaction) as a function of size
- different types (metallic, van-der-Waals, etc.); size dependent
- easily produced (with size distribution); size-selection in collision physics

bridge between { atoms (finite systems) / solids (high density) } but unique properties

e. g. very strong charging → explosion

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

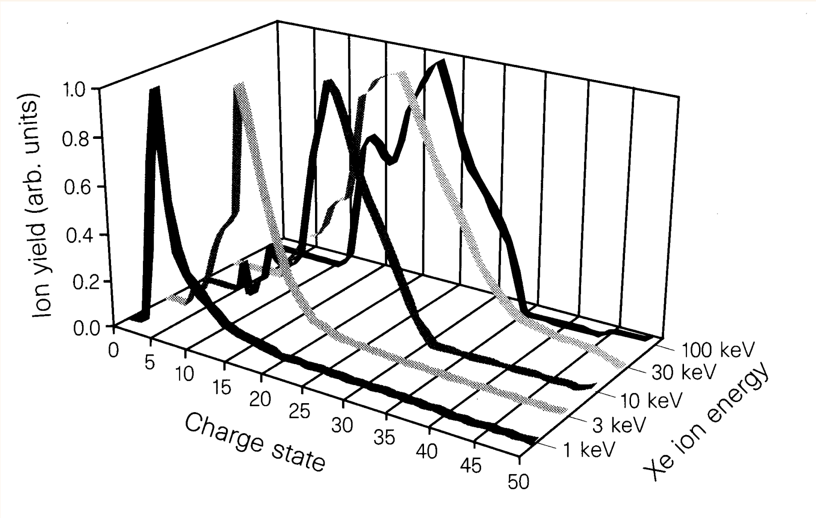
neutrons

experiments @ 780 nm

Ditmire et al. 1997:

up to Xe^{35+} with 100 keV

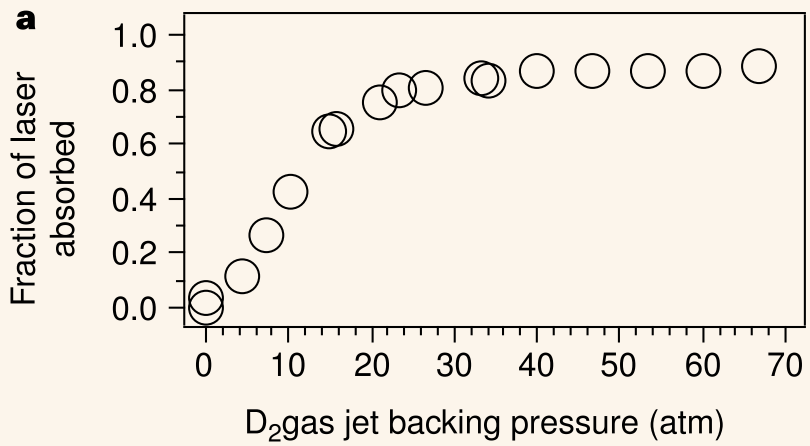
from $Xe_{20.000}$



Ditmire et al. 1999:

very efficient absorption

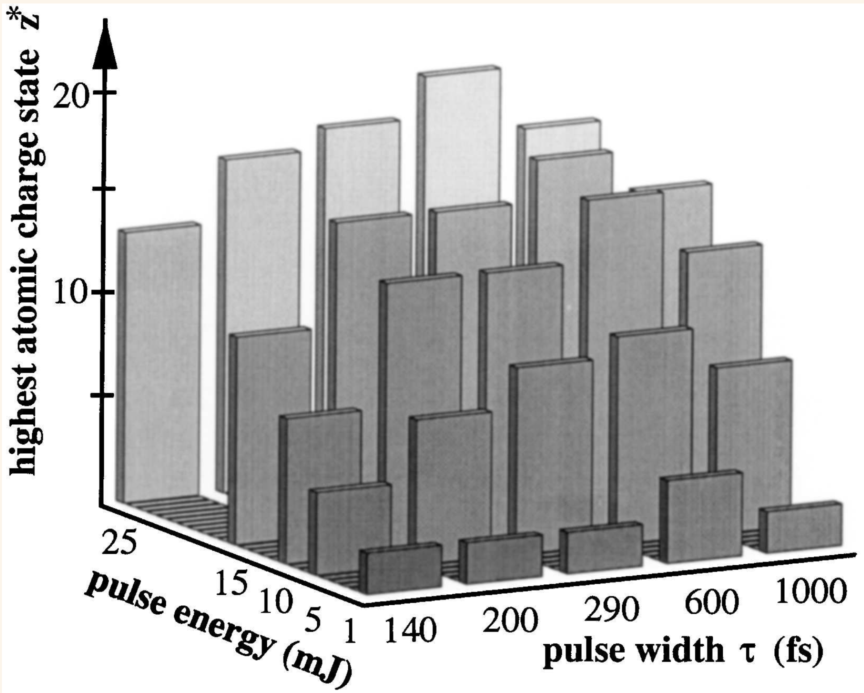
nuclear fusion of fragments



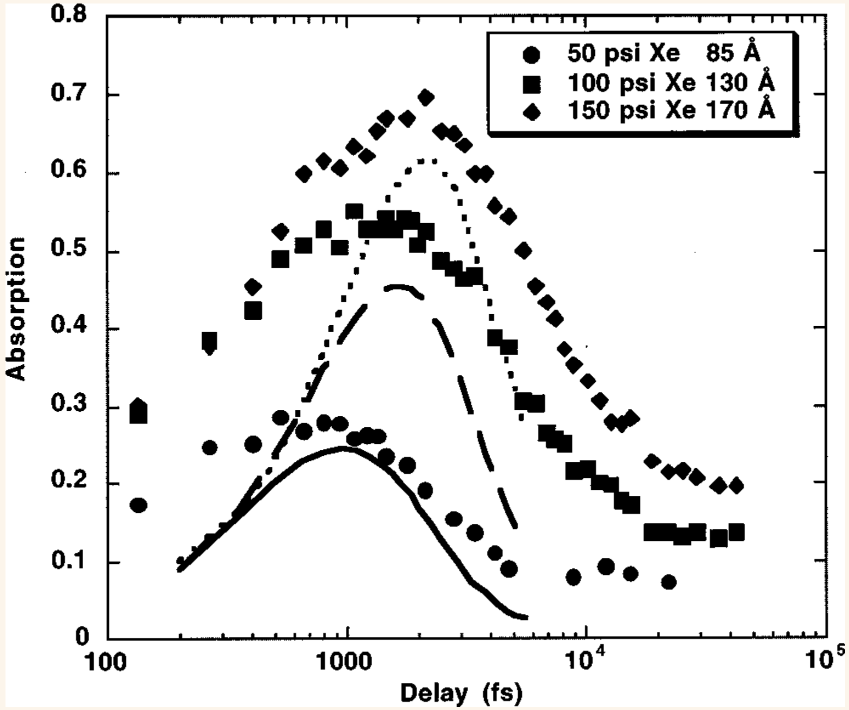
higher charge states and much faster fragments than for molecules !

experiments @ 780 nm

platinum: Meiwes-Broer et al. 1999
variable pulse length (fixed fluence)



xenon: Ditmire et al. 1999
pump-probe measurement

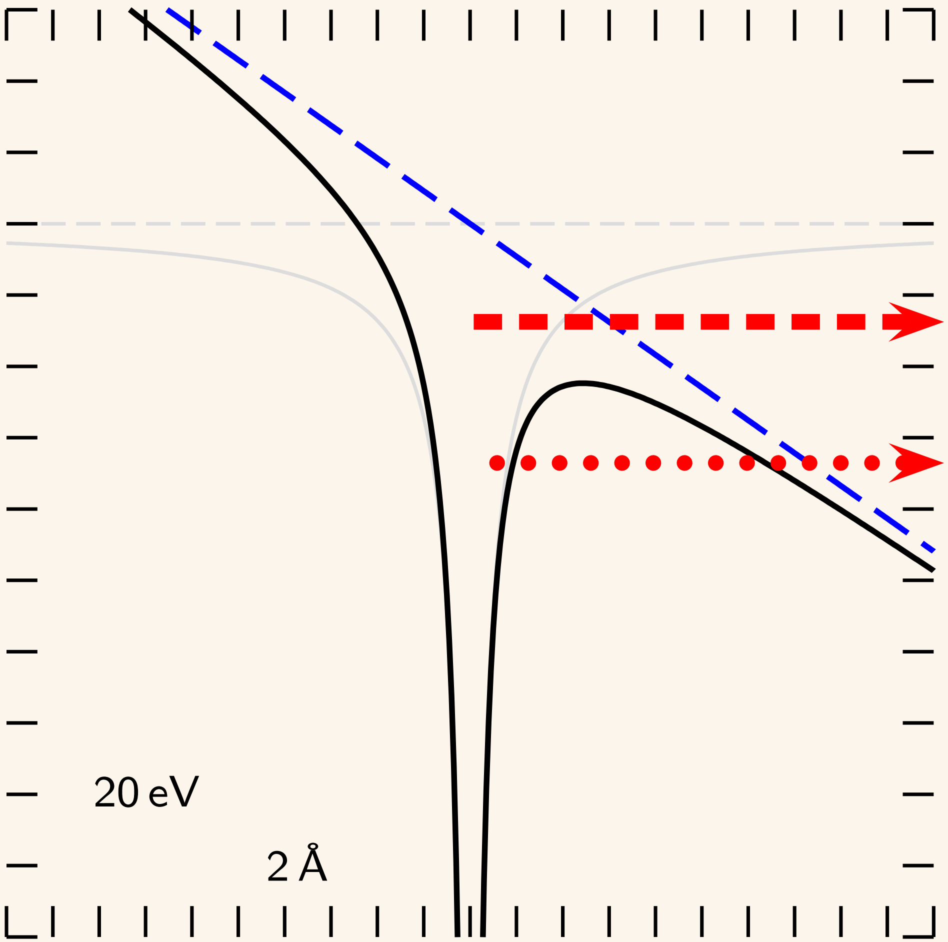


dependence on pulse length / pump-probe delay at a femtosecond time scale

atoms in strong fields

$$\hat{H} = \frac{1}{2}\left(\hat{\vec{p}} - e\vec{A}\right)^2 + V(\vec{r}) \quad \rightarrow \quad \frac{1}{2}\hat{\vec{p}}^{\,2} + V(\vec{r}) \; - \; e\frac{\vec{\mathcal{E}}}{\omega}\hat{\vec{p}} + \underbrace{\frac{e^2}{2}\frac{\vec{\mathcal{E}}^{\,2}}{\omega^2}}_{}$$

$$2E_{\text{pond}} \leftrightarrow E_{\text{bind}}$$



Keldysh parameter

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

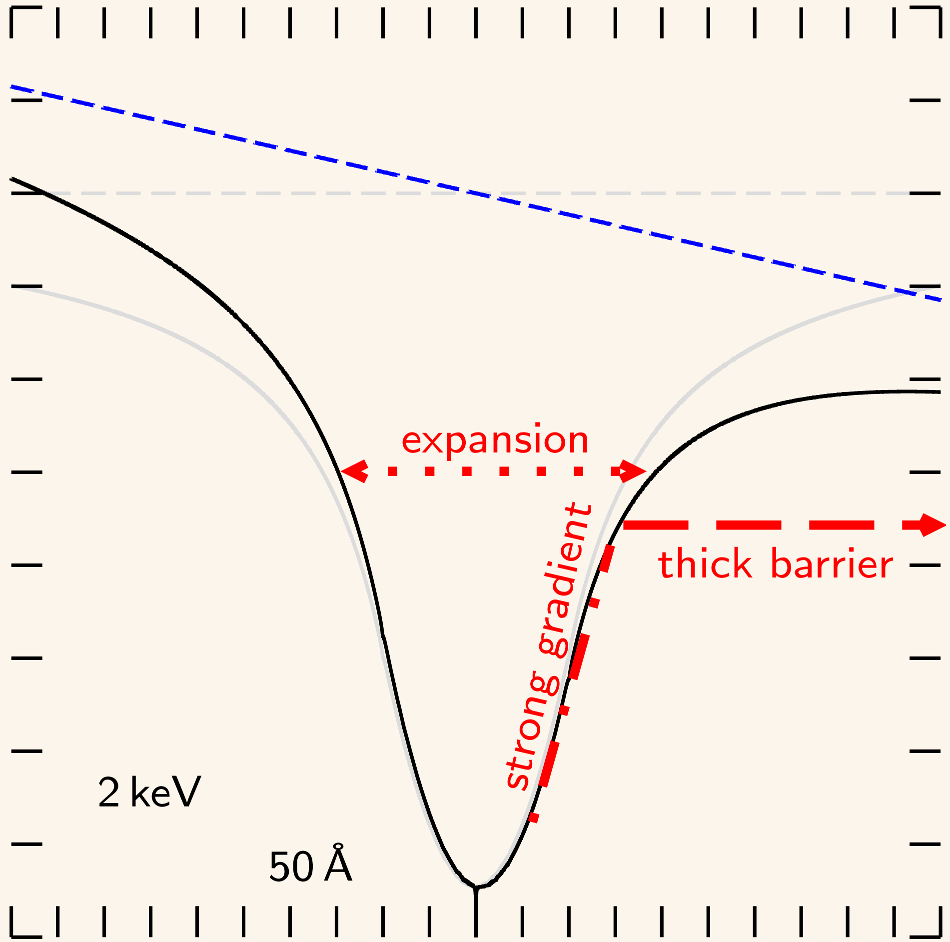
$\gamma < 1$ **field ionization**

(over-the-barrier or tunnel)

clusters vs. atoms in strong fields

atom

cluster: 10^4 atoms



$$V(r) = \begin{cases} -\dfrac{3R^2 - r^2}{2R^2}\dfrac{Q}{R} & r \le R \\ -\dfrac{Q}{r} & r \ge R \end{cases}$$

depth $\sim \dfrac{Q}{R} \sim R^2$ gradient $\sim \dfrac{Q}{R^2} \sim R$

treatment of laser-cluster interaction

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

two-step approach

- atomic (inner) ionization:

  bound electrons $\longrightarrow$ "quasi-free" electrons

- cluster (outer) ionization:

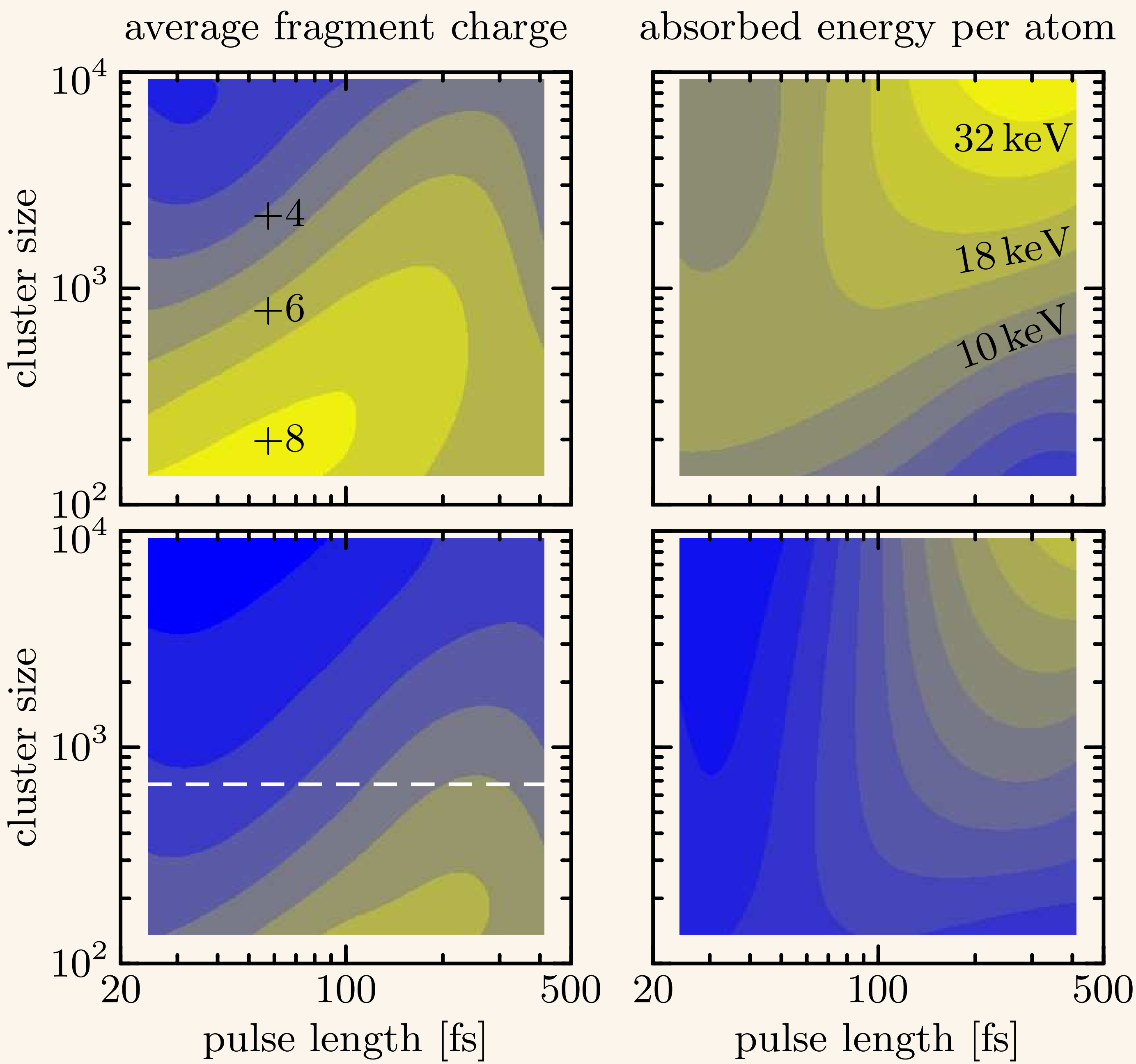
  "quasi-free" electrons $\longrightarrow$ free electrons

Rose-Petruck et al. 1995

treatment of laser-cluster interaction

- 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



pulses with fixed fluence

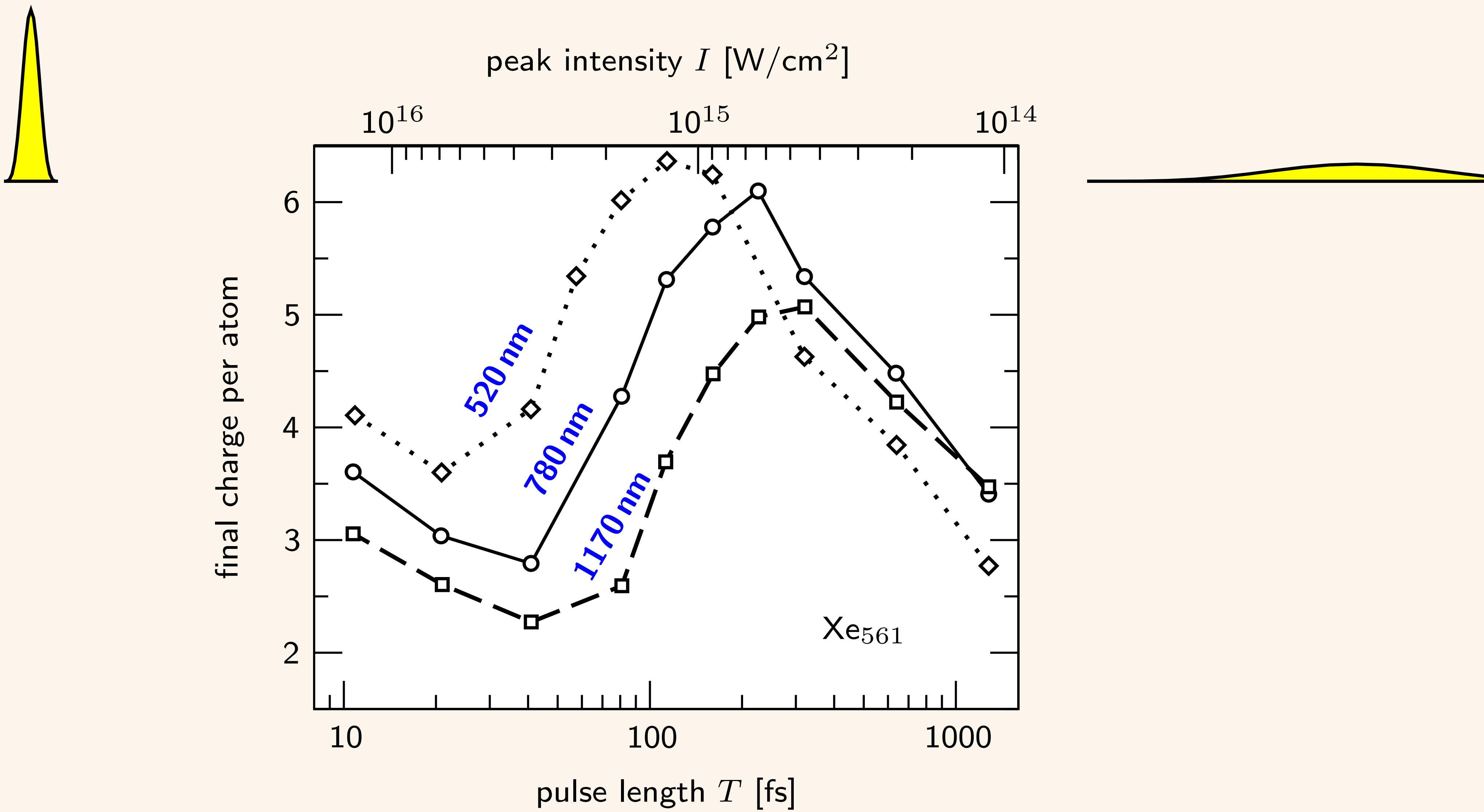
2×10^{16} W/cm^2 @ 100 fs

4×10^{15} W/cm^2 @ 100 fs

pulse-length dependence for different wavelengths

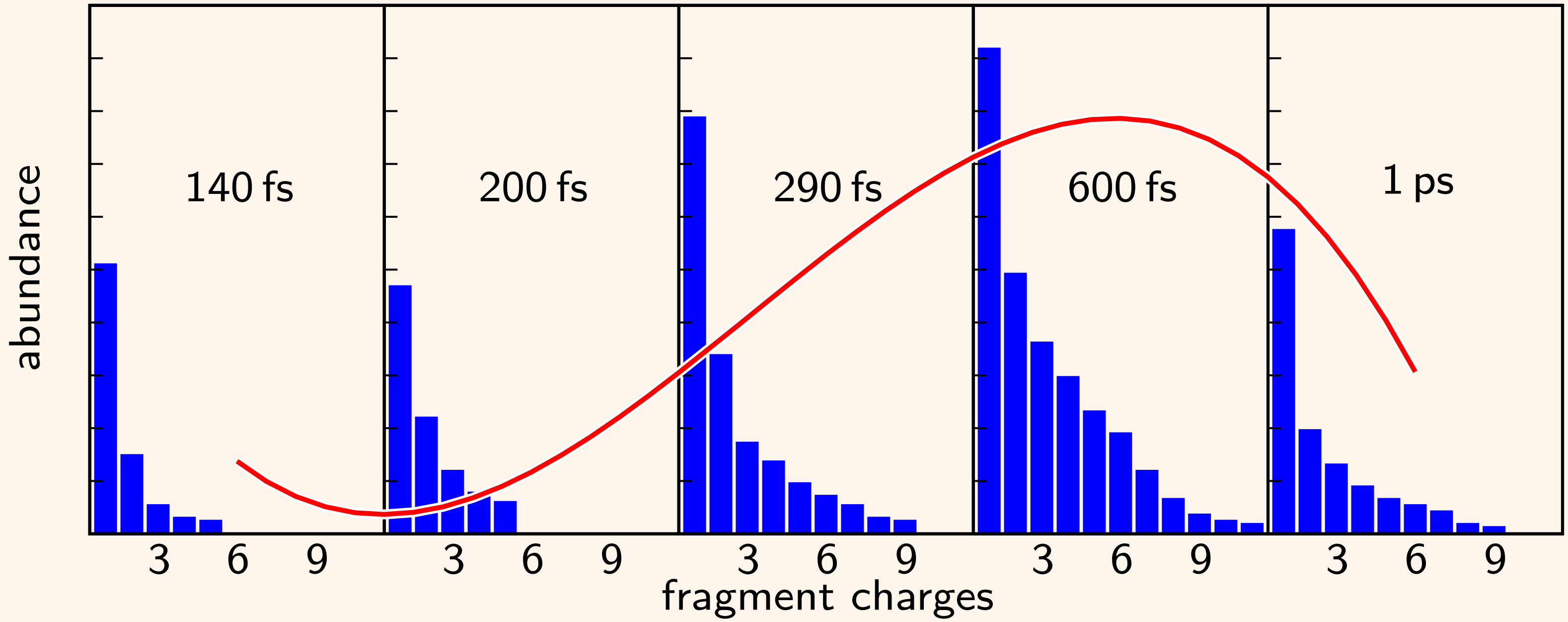
[US & Rost, PRL 91 (2003) 223401]

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



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 $F(t) = F_0 \cos(\omega t)$

$\downarrow$

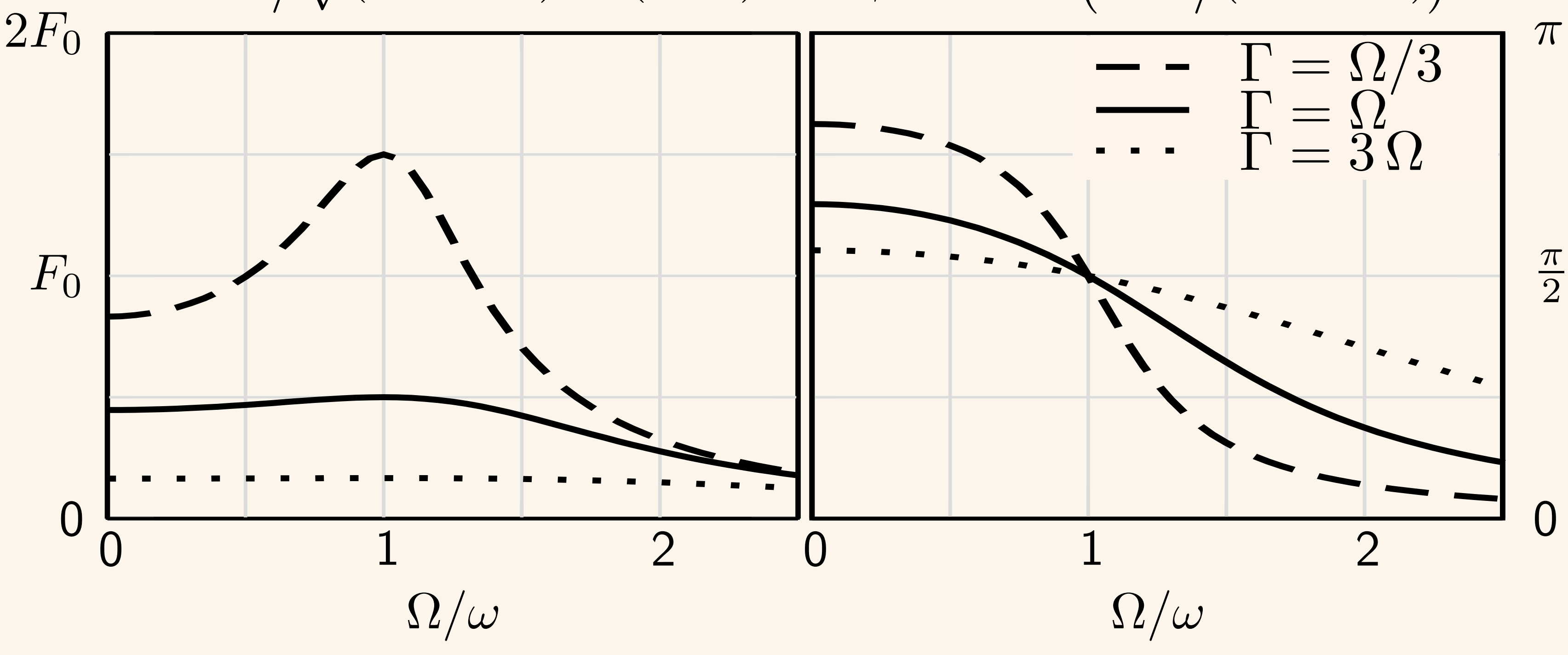
$$X(t) = \mathbf{A_t} \cos(\omega t - \phi_{\mathbf{t}})$$

amplitude

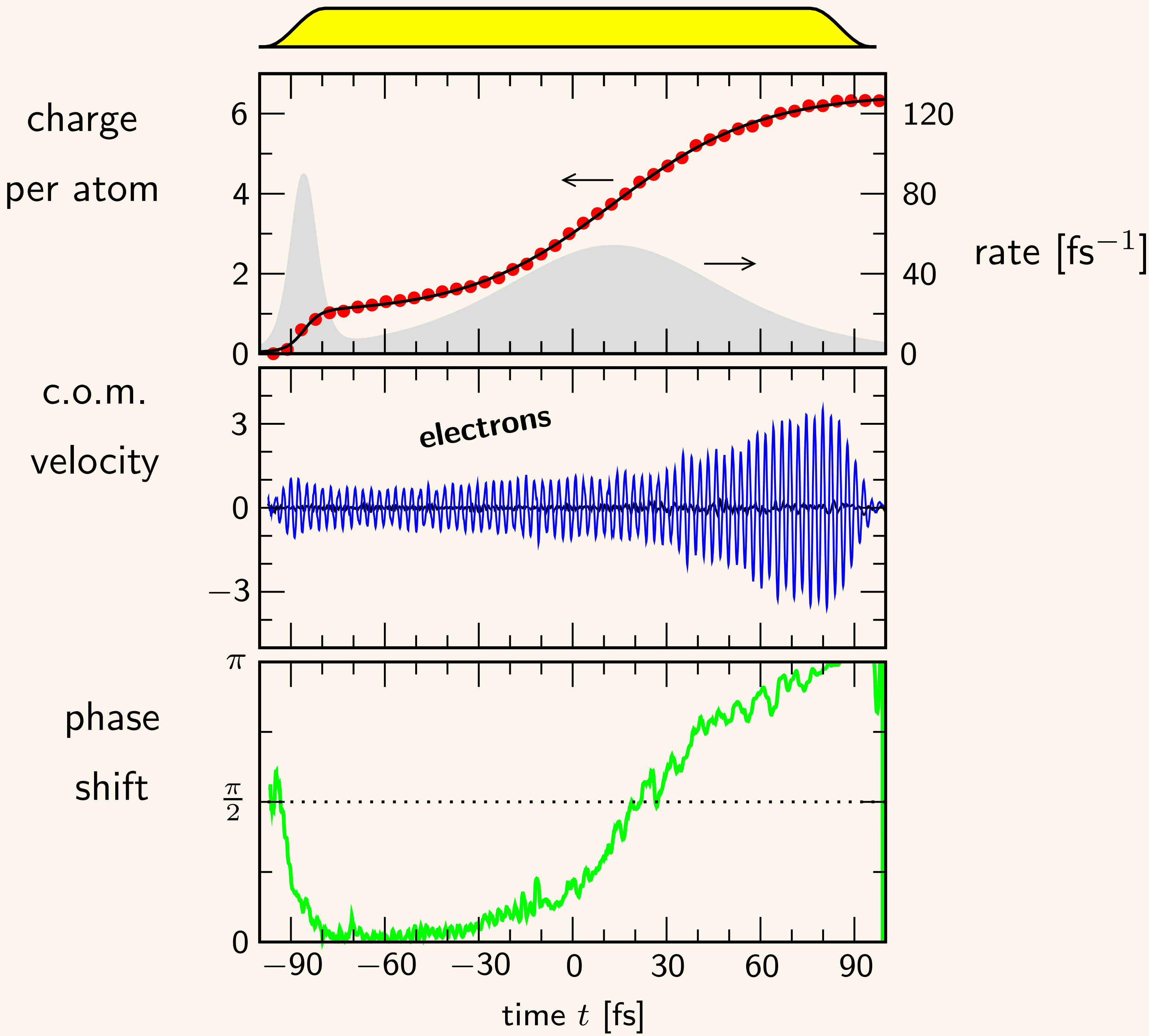
$$A = F_0 \Big/ \sqrt{({\Omega_t}^2 - \omega^2)^2 + (2\Gamma_t \omega)^2}$$

phase

$$\phi = \arctan\left(2\Gamma_t \omega \big/ ({\Omega_t}^2 - \omega^2)\right)$$



cluster dynamics $(Xe_{923} @ 9\times10^{14}\,W/cm^2)$

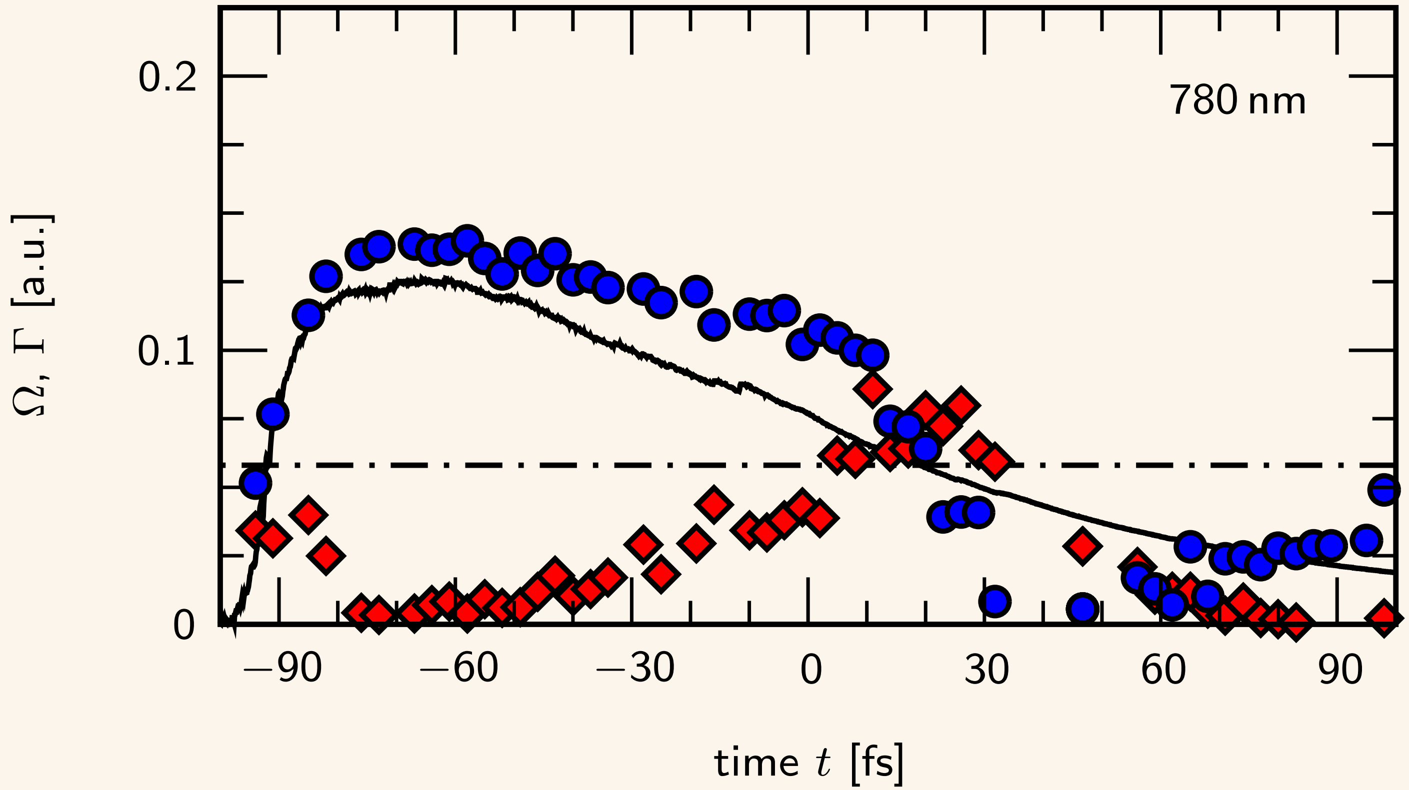


simple model vs. many-particle dynamics

$$\Omega_t{}^2 = \omega^2 + (F_0/A_t)\cos\phi_t$$

$$\Gamma_t = \big(F_0/(2A_t\omega)\big)\sin\phi_t$$

$$\Omega_t = \sqrt{Q_{\rm ion}(t)/R(t)^3}$$



nano-plasma vs. harmonic oscillator model

[Ditmire et al. 1995]

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

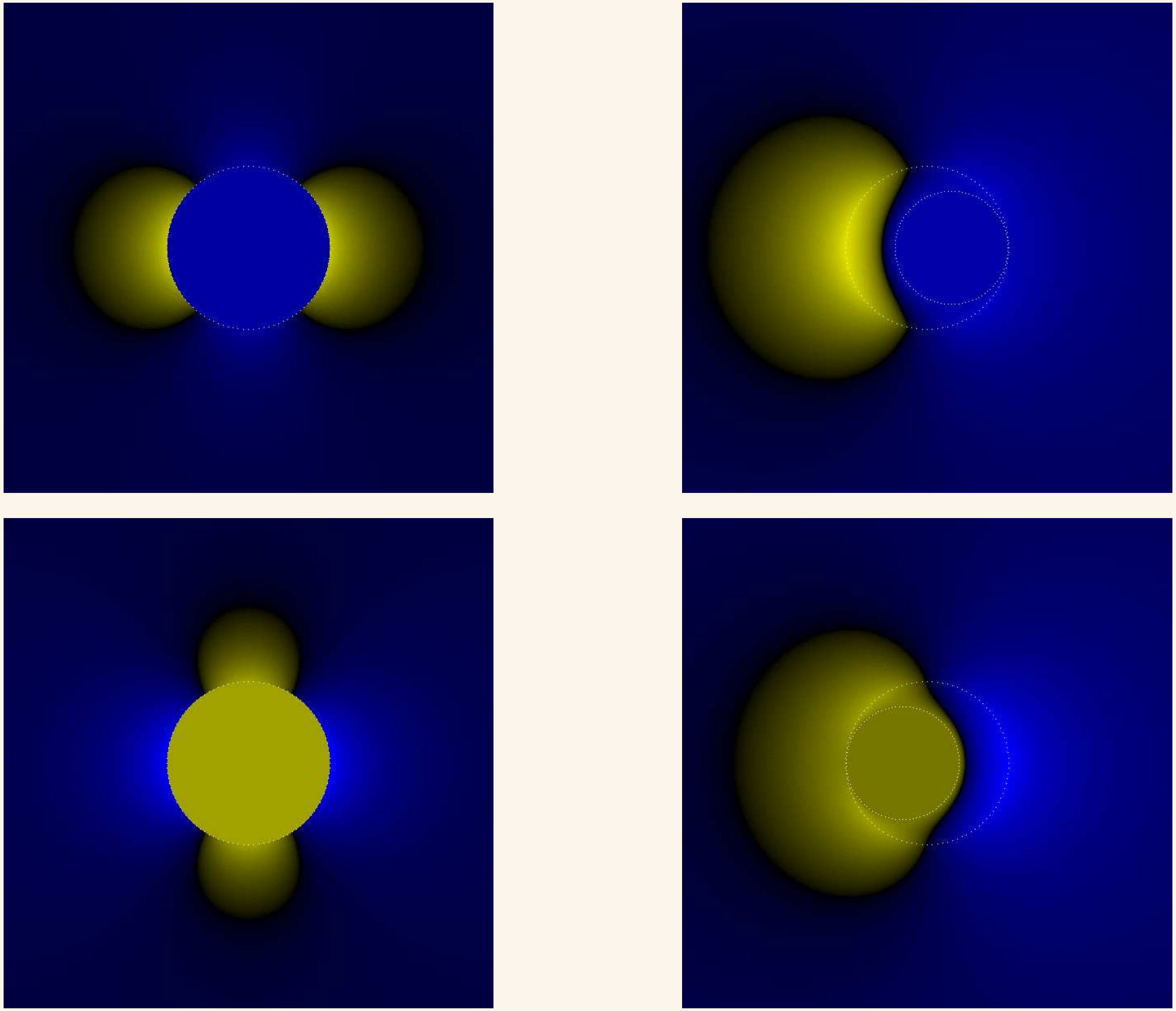
critical density $\omega_{\mathrm{crit}} = \sqrt{\dfrac{4\pi\rho}{3}} = \dfrac{\omega_{\mathrm{p}}}{\sqrt{3}}$

harmonic oscillator model

eigenfrequency $\Omega_t = \sqrt{\dfrac{Q_{\mathrm{ion}}}{R^3}} = \sqrt{\dfrac{4\pi\rho_{\mathrm{ion}}}{3}}$

nano-plasma vs. harmonic oscillator model

electric field | nano-plasma | harmonic oscillator



below resonance

$\Omega < \omega$

above resonance

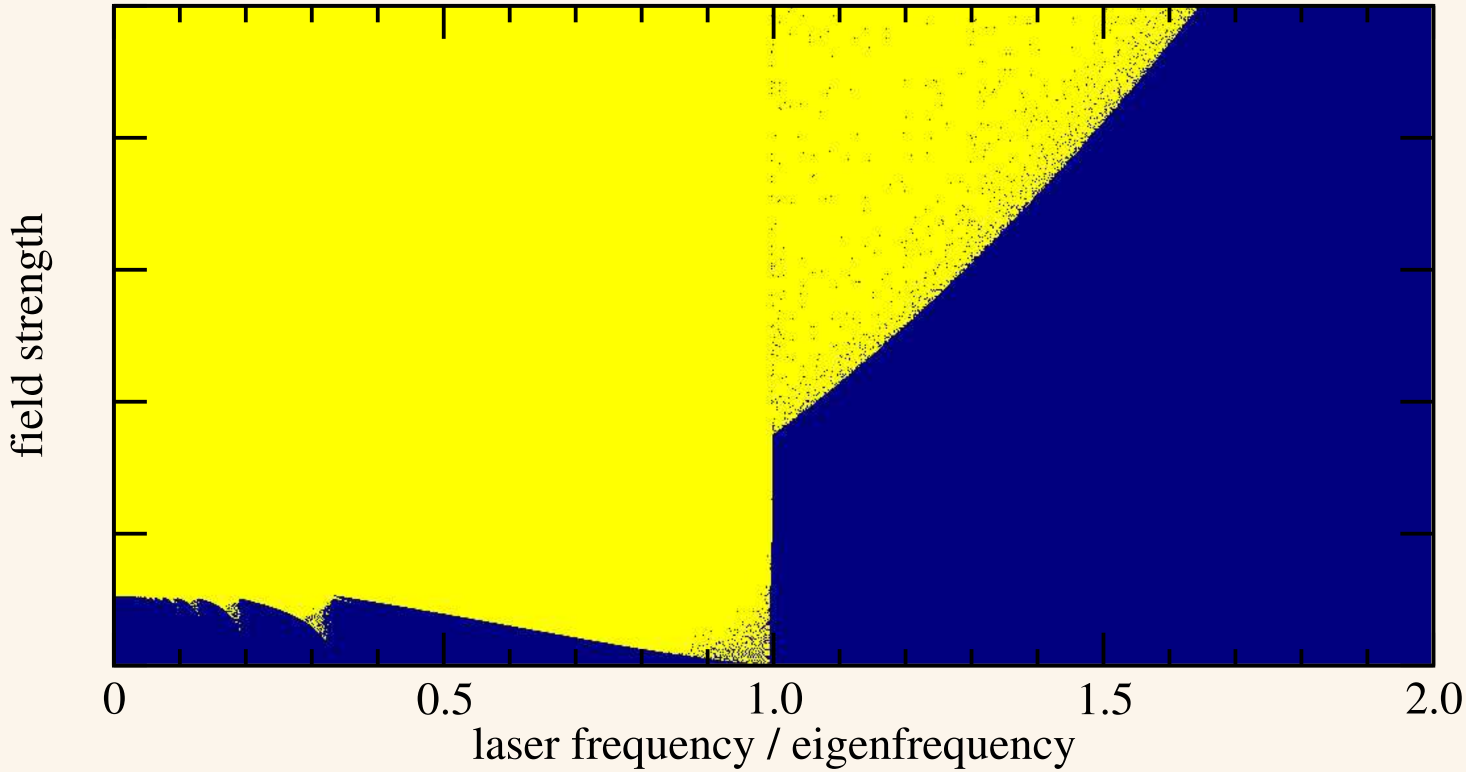
$\Omega > \omega$

negative ↑ positive

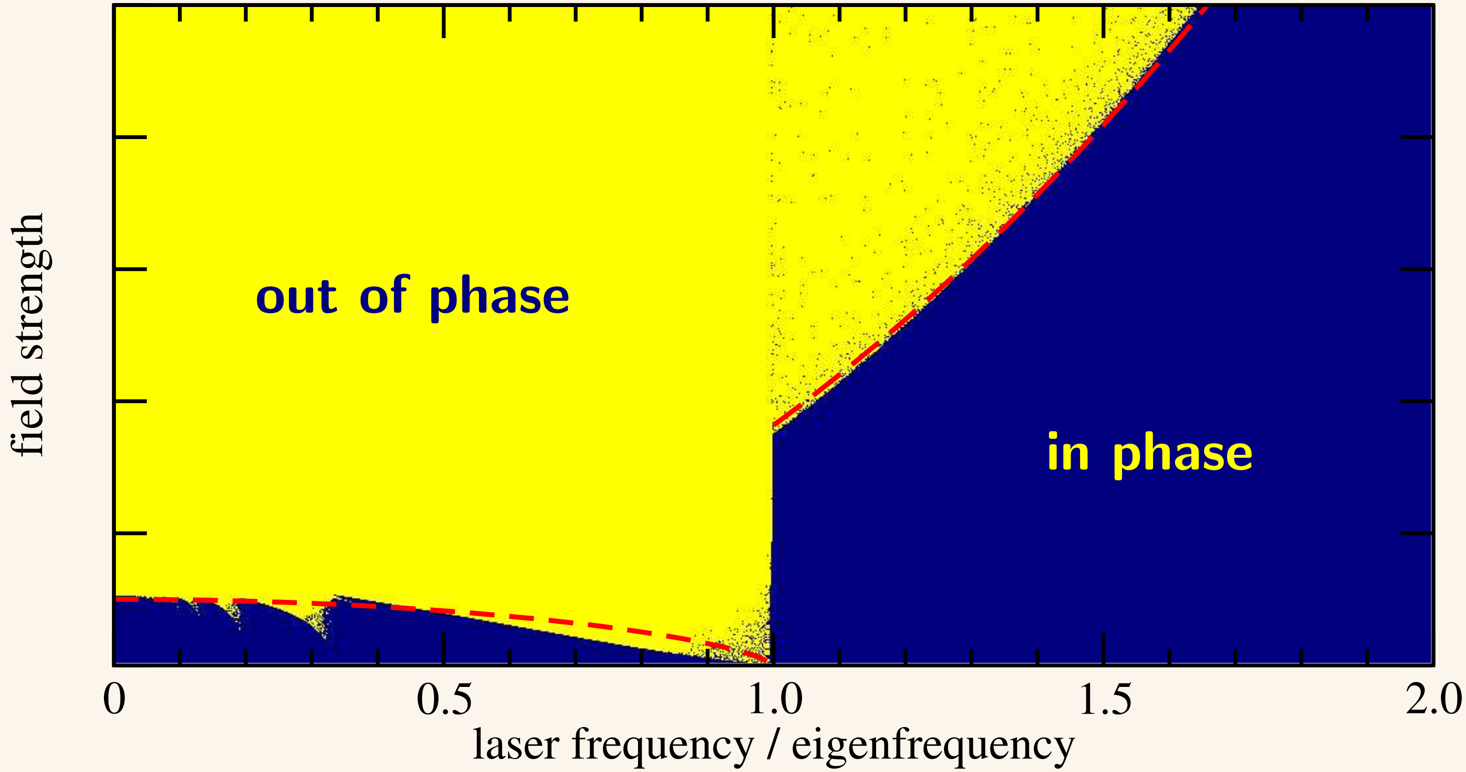
ionization in a rigid cloud model

Gaussian ion and electron clouds $\longrightarrow$ interaction potential

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



ionization in a rigid cloud model



cycle-averaged potential (KH frame)

$$\overline{V}(r) = \int_{-\pi}^{+\pi} \mathrm{d}\phi \, V(|r - \alpha_0 \sin(\phi)|)$$

becomes a double well

$$\frac{\mathrm{d}^2}{\mathrm{d}r^2}\overline{V}(r)\bigg|_{r=0} = 2\int_0^{\pi} \mathrm{d}\phi \, V''(\alpha_0 \sin(\phi)) < 0$$

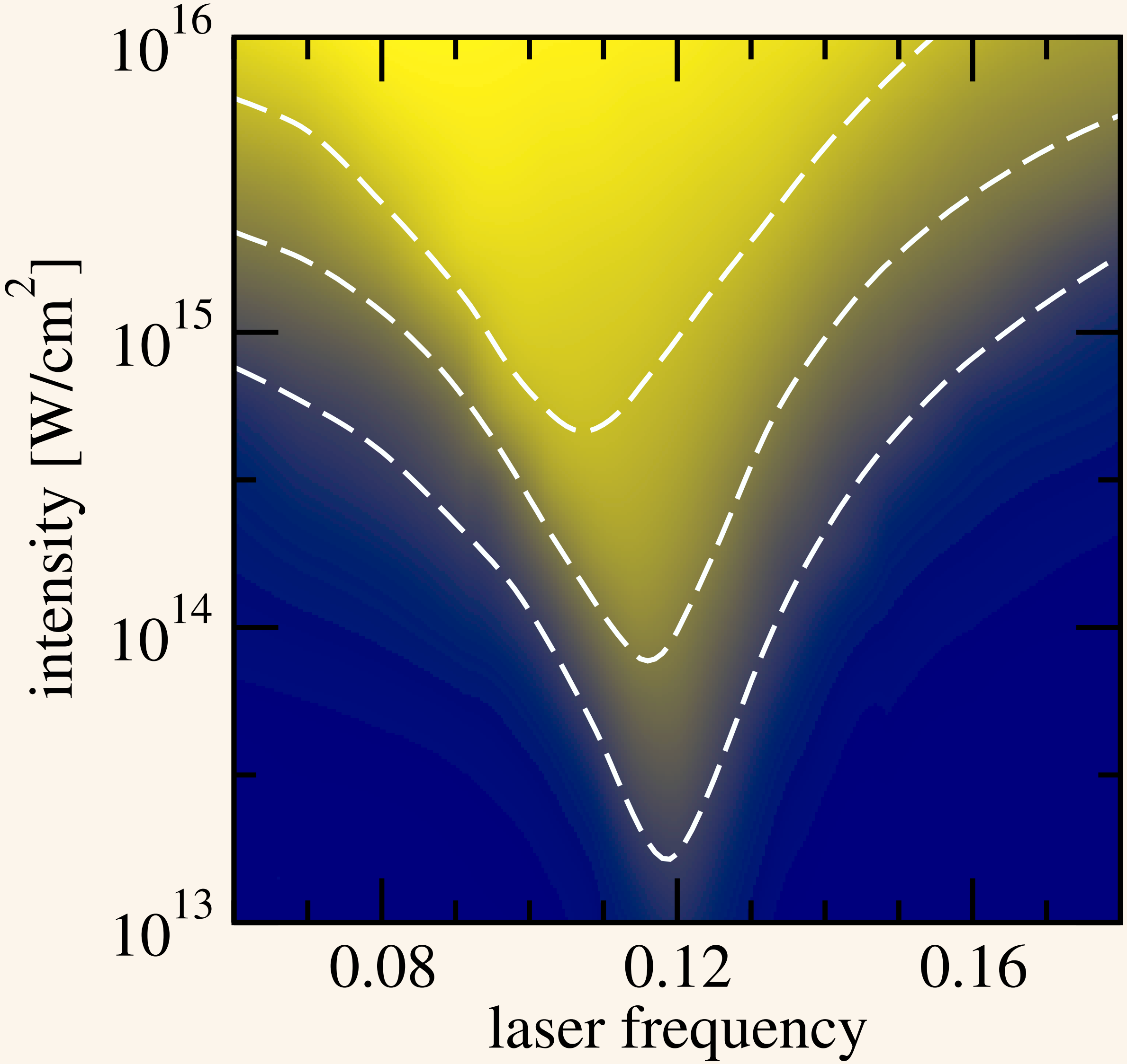
$$\rightarrow \alpha_0 \approx 2R \qquad \text{or} \qquad F(w) \approx 2R\omega^2$$

amplitude (harmonic) = barrier (Coulombic)

$$a = \frac{F}{\Omega^2 - \omega^2} \qquad b = \sqrt{\frac{Q}{F}}$$

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

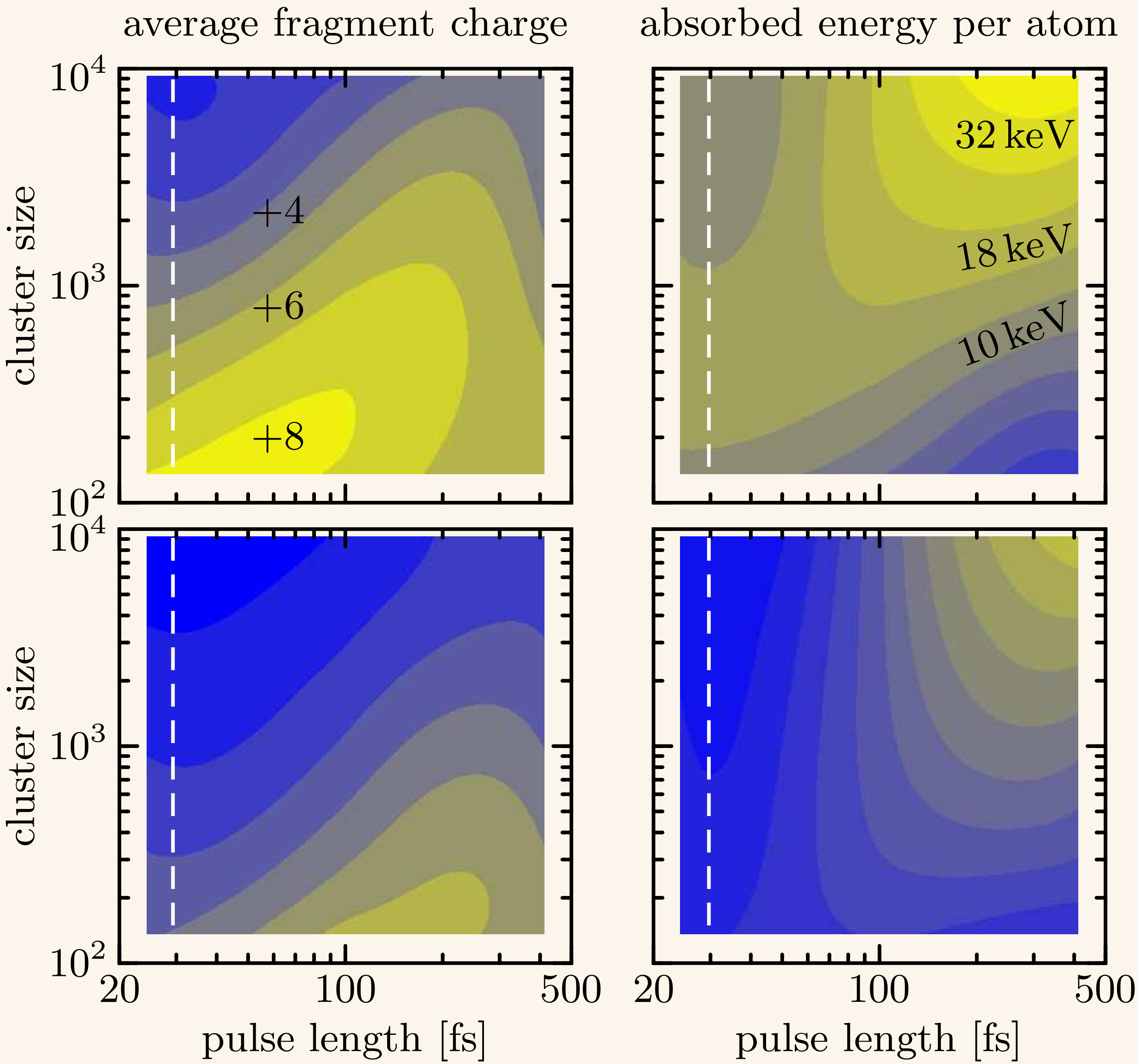
ionization in a many-electron model



ionization probability

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

cluster-size and pulse-length dependence @ 780 nm



pulses with fixed fluence

2×10^{16} W/cm^2 @ 100 fs

4×10^{15} W/cm^2 @ 100 fs

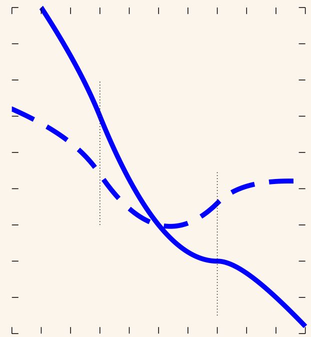
ionization in ultra-short pulses

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

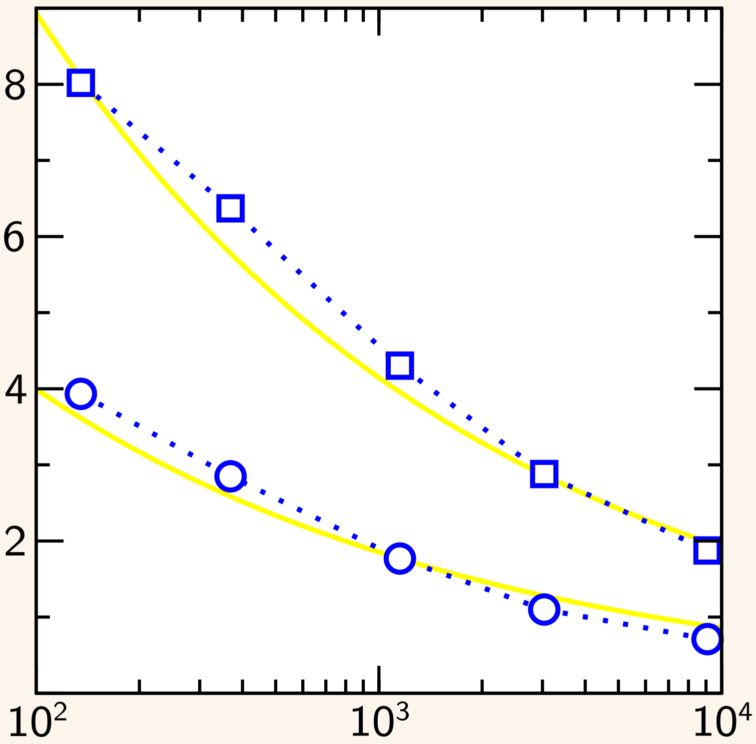
at $I = \frac{16}{3.2} \times 10^{15}$ W/cm^2

field ionization model

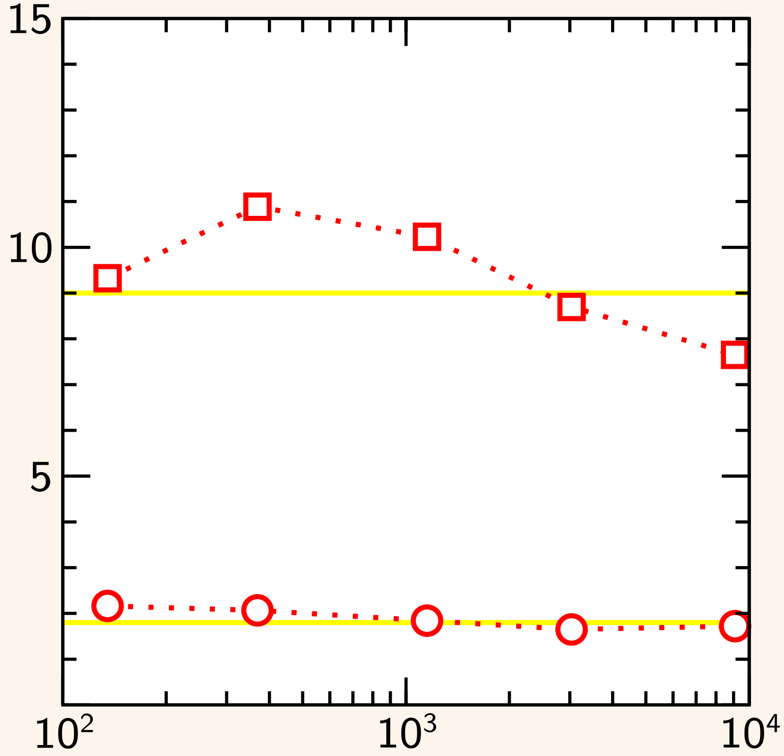
$q \propto \sqrt{I}/R \qquad E \propto I$



average fragment charges



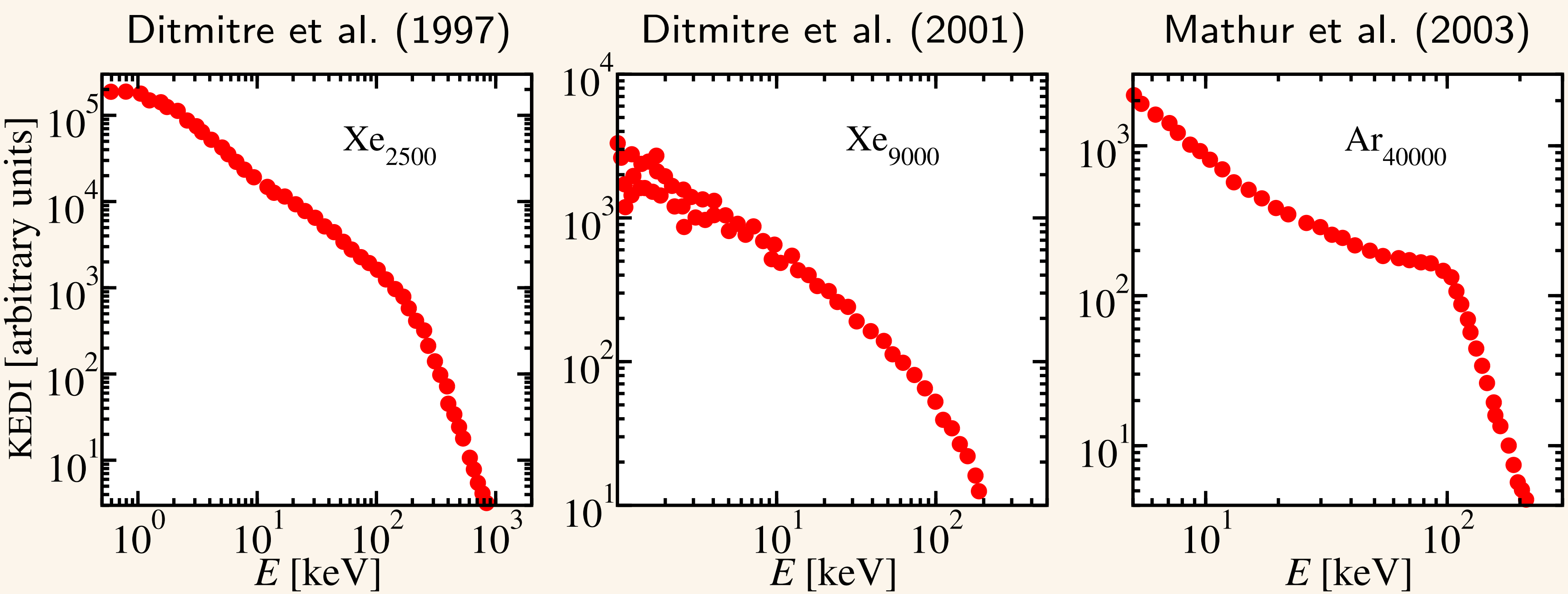
absorbed energy per atom



kinetic energy distribution of ions

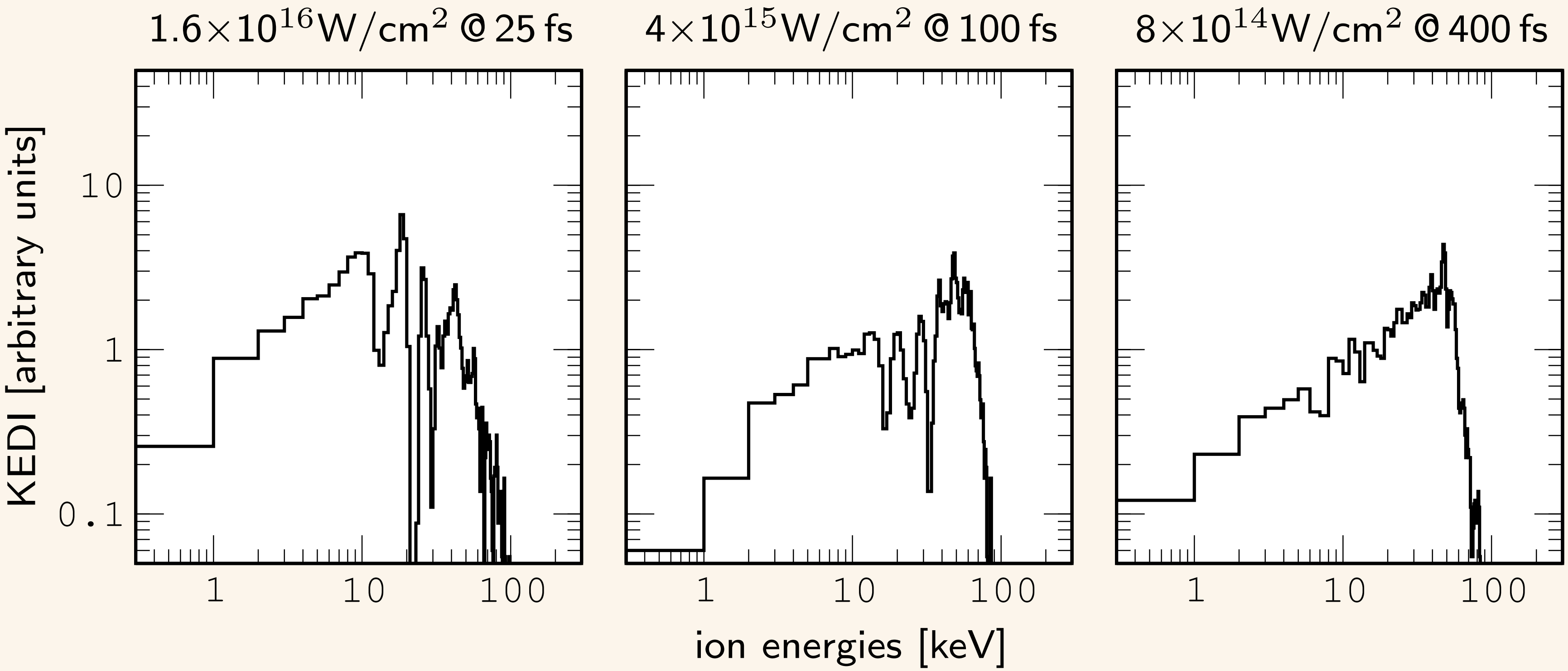
"standard" experimental observable

data from different groups studying different targets



kinetic energy distribution of ions

microscopic single-cluster calculations for Xe_{9093}



kinetic energy distribution of ions

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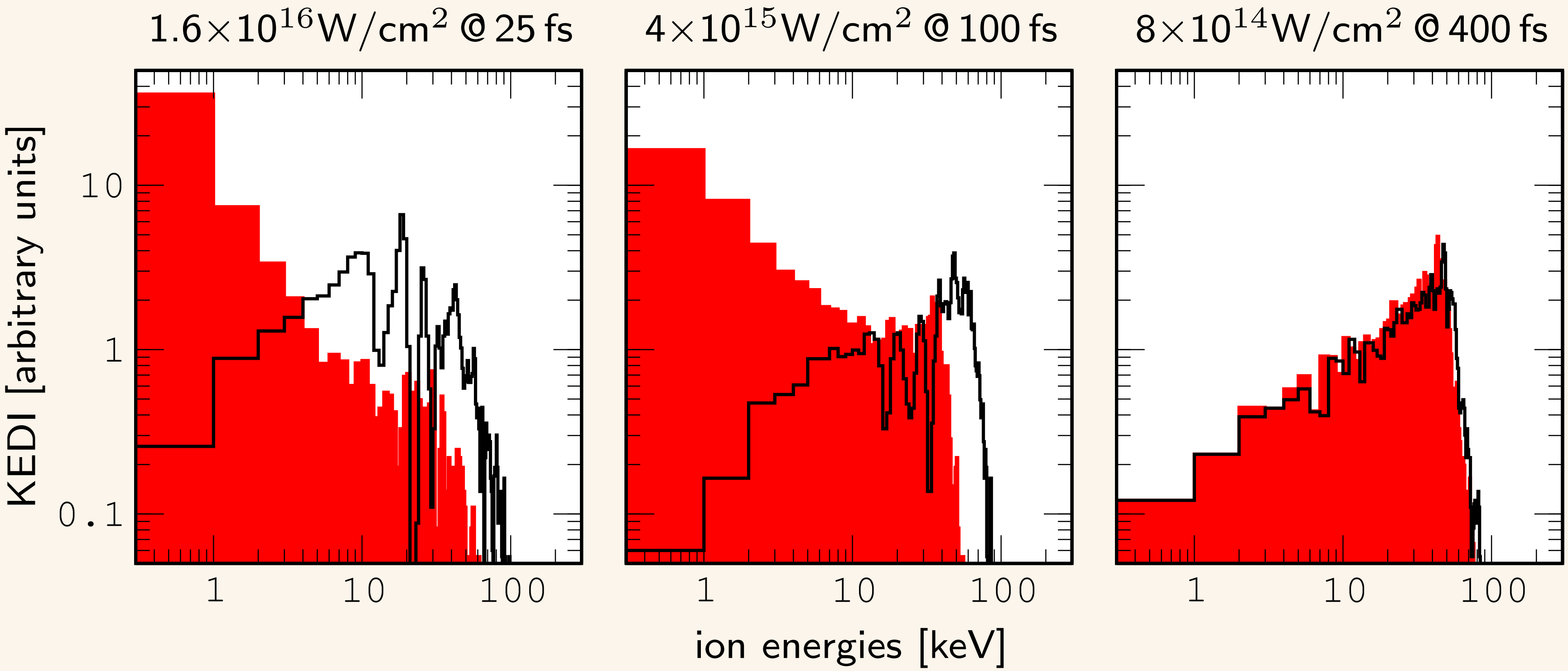
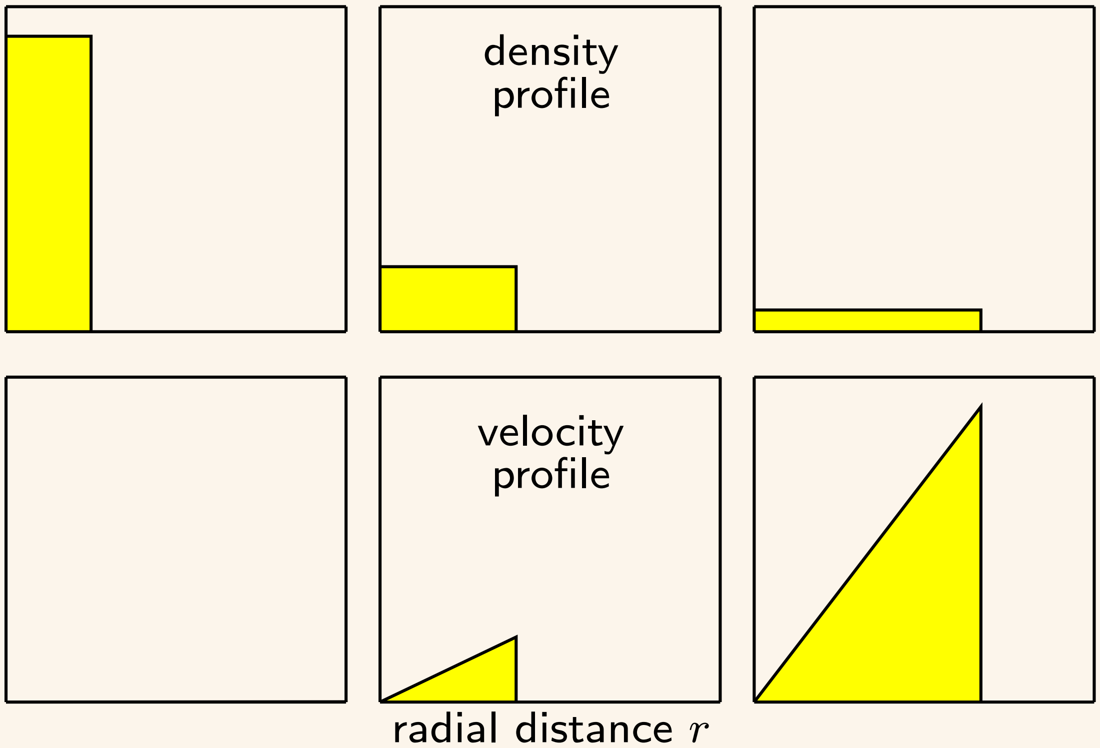


kinetic energy distribution of ions

simple model of a expanding charged sphere

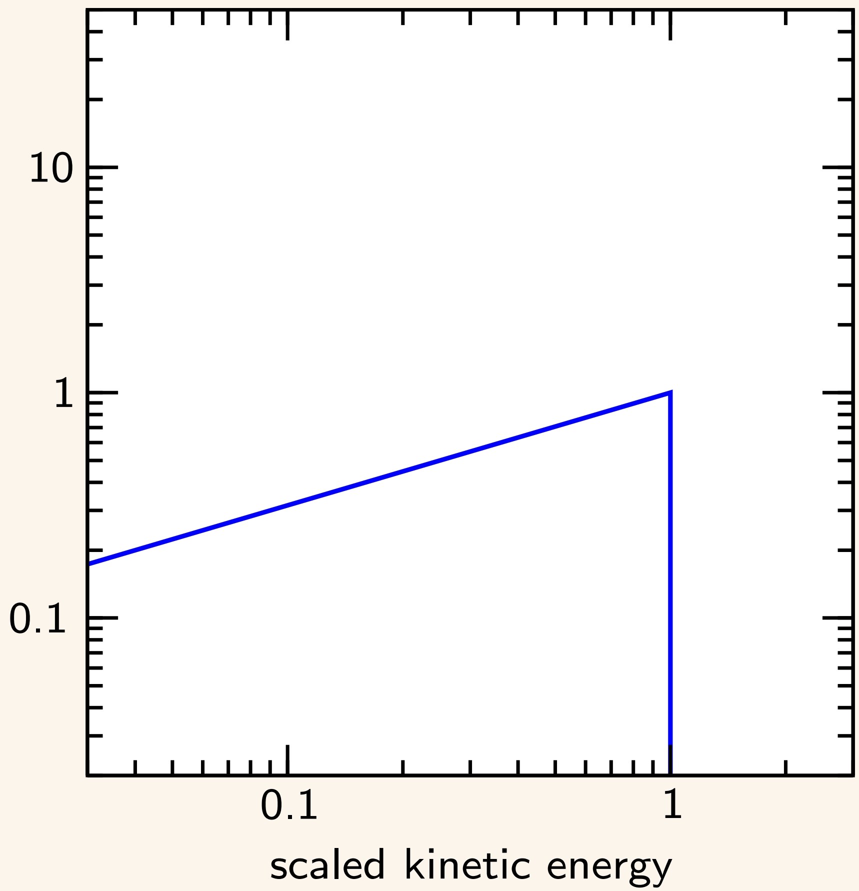
shell at radial distance r: energy $E \propto r^4 \rightarrow$ field $F \propto r^3 \rightarrow$ acceleration $a \propto r$

$\rightarrow$ cluster keeps its shape



kinetic energy distribution of ions

single cluster in a homogeneous field



homogeneous charge distribution in the cluster

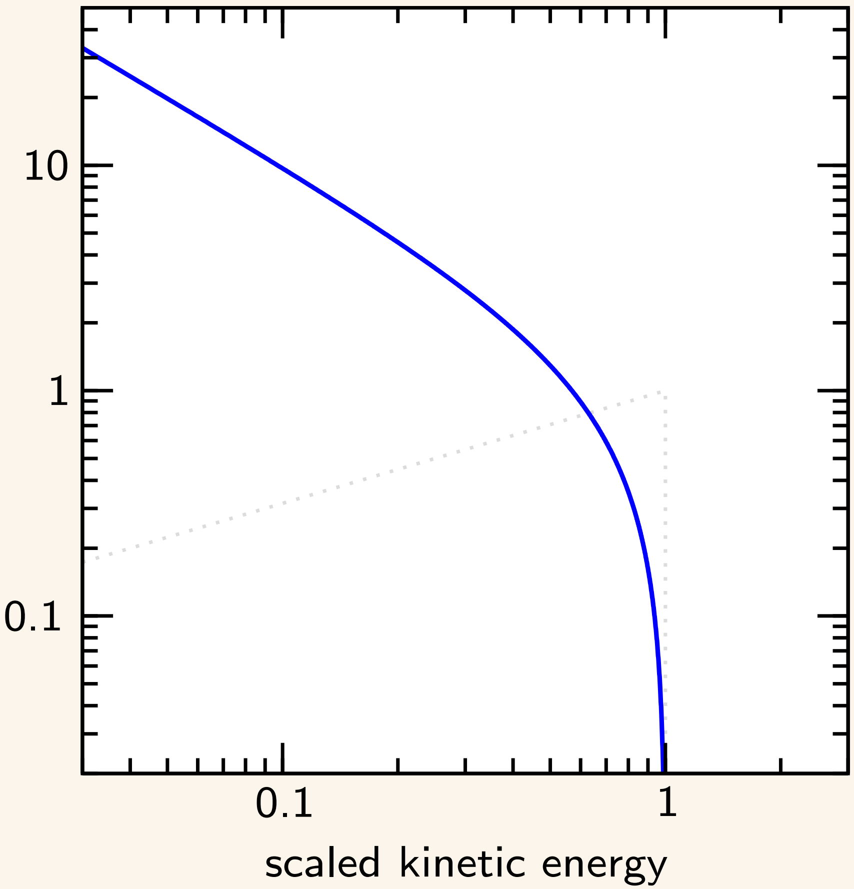
$$\frac{dP}{d\varepsilon} = \frac{3}{2}\sqrt{\varepsilon}\,\Theta(1-\varepsilon)$$

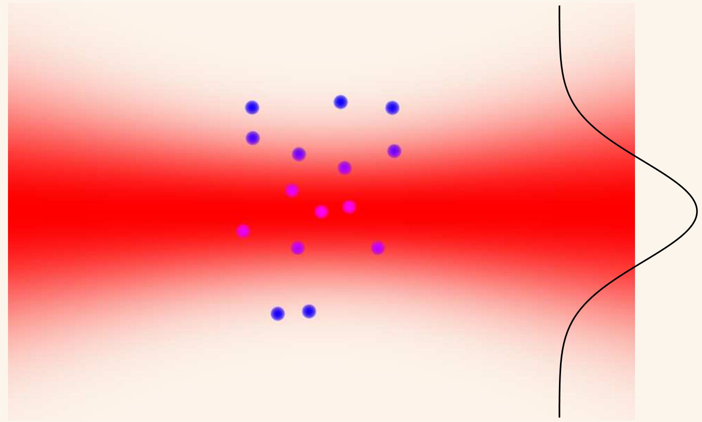
fastest ions from the surface

Note: scaled energies !

kinetic energy distribution of ions

clusters from Gaussian laser focus





folding with laser intensity

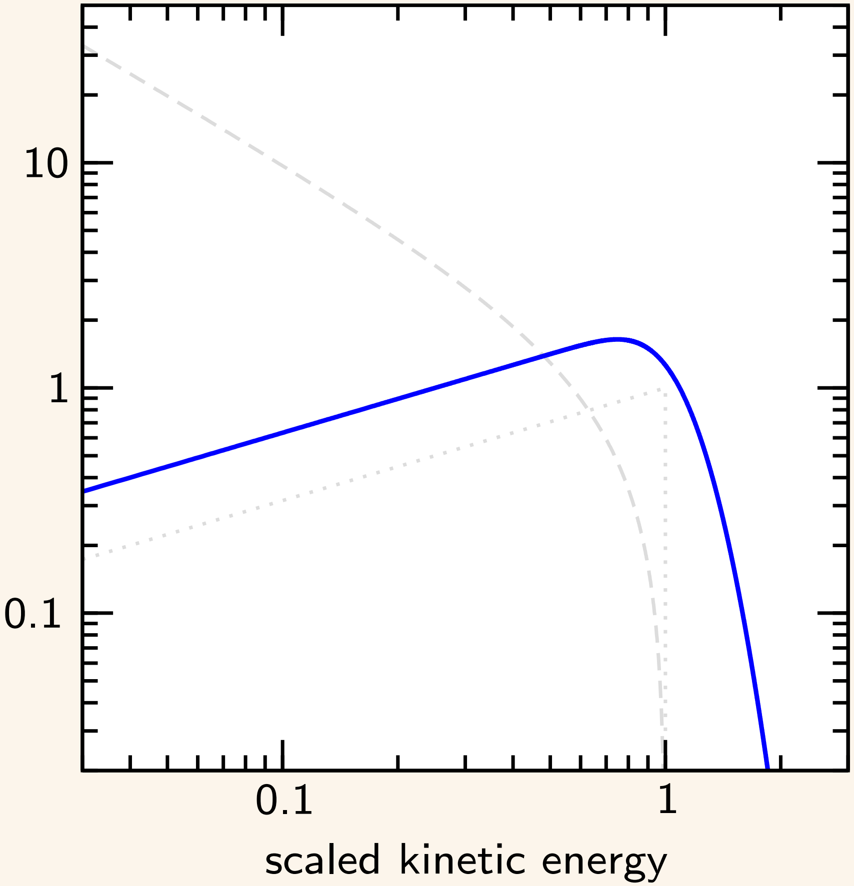
$$\sim \exp\left(-r^2/\xi^2\right)$$

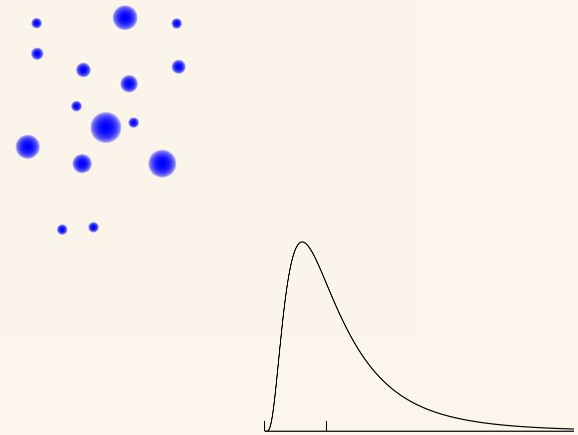
$$\frac{dP_{\mathrm{las}}}{d\varepsilon} = \frac{\pi\xi^2 N}{2} \frac{1-\varepsilon^{3/2}}{\varepsilon} \Theta(1-\varepsilon)$$

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

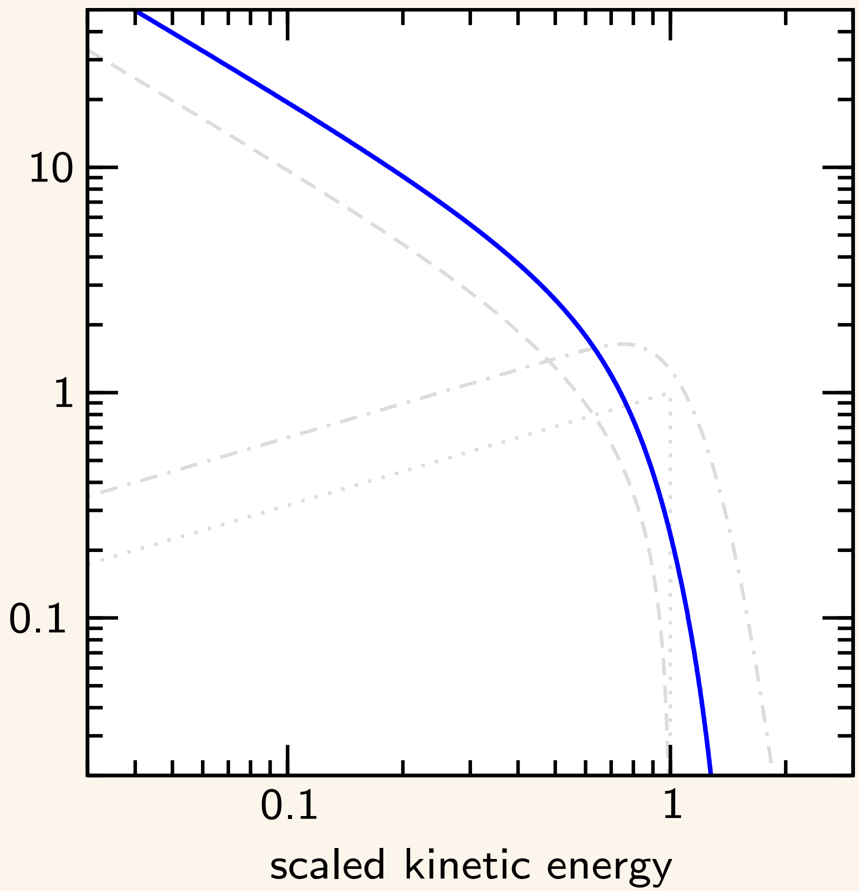
$$\sim \frac{1}{N} \exp\left(-\frac{\ln^2(N/N_0)}{2\nu^2}\right)$$

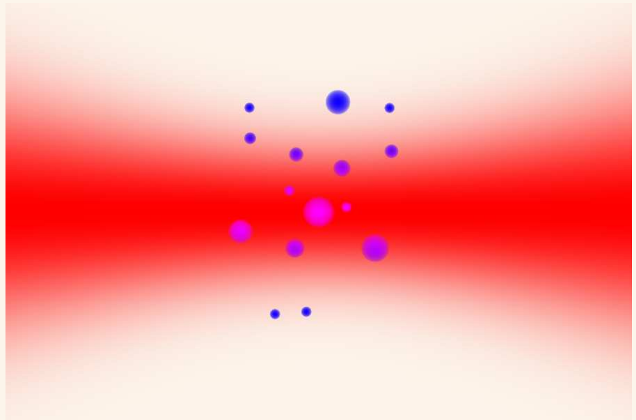
$$\frac{dP_{\text{size}}}{d\varepsilon} = \frac{3}{4} N_0 \sqrt{\varepsilon}\, \text{erfc}\left(\frac{\frac{3}{2}\ln\varepsilon}{\sqrt{2}\nu}\right)$$

fastest ions from larger clusters

kinetic energy distribution of ions

cluster size distribution from Gaussian laser





folding with both distributions

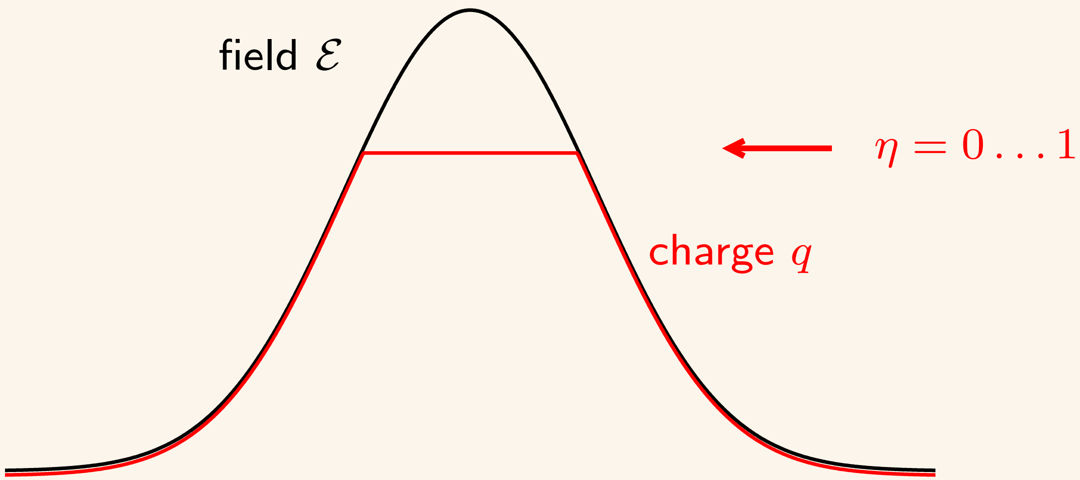
$$\frac{dP_{\text{both}}}{d\varepsilon} = \frac{\xi^2\pi}{4}\frac{N_0}{\varepsilon}\left[\varepsilon^{3/2}\text{erfc}\left(\frac{3\ln\varepsilon}{2\sqrt{2}\nu}\right) + e^{\nu^2/2}\left(1+\text{erf}\left(\frac{2\nu^2-3\ln\varepsilon}{2\sqrt{2}\nu}\right)\right)\right]$$

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

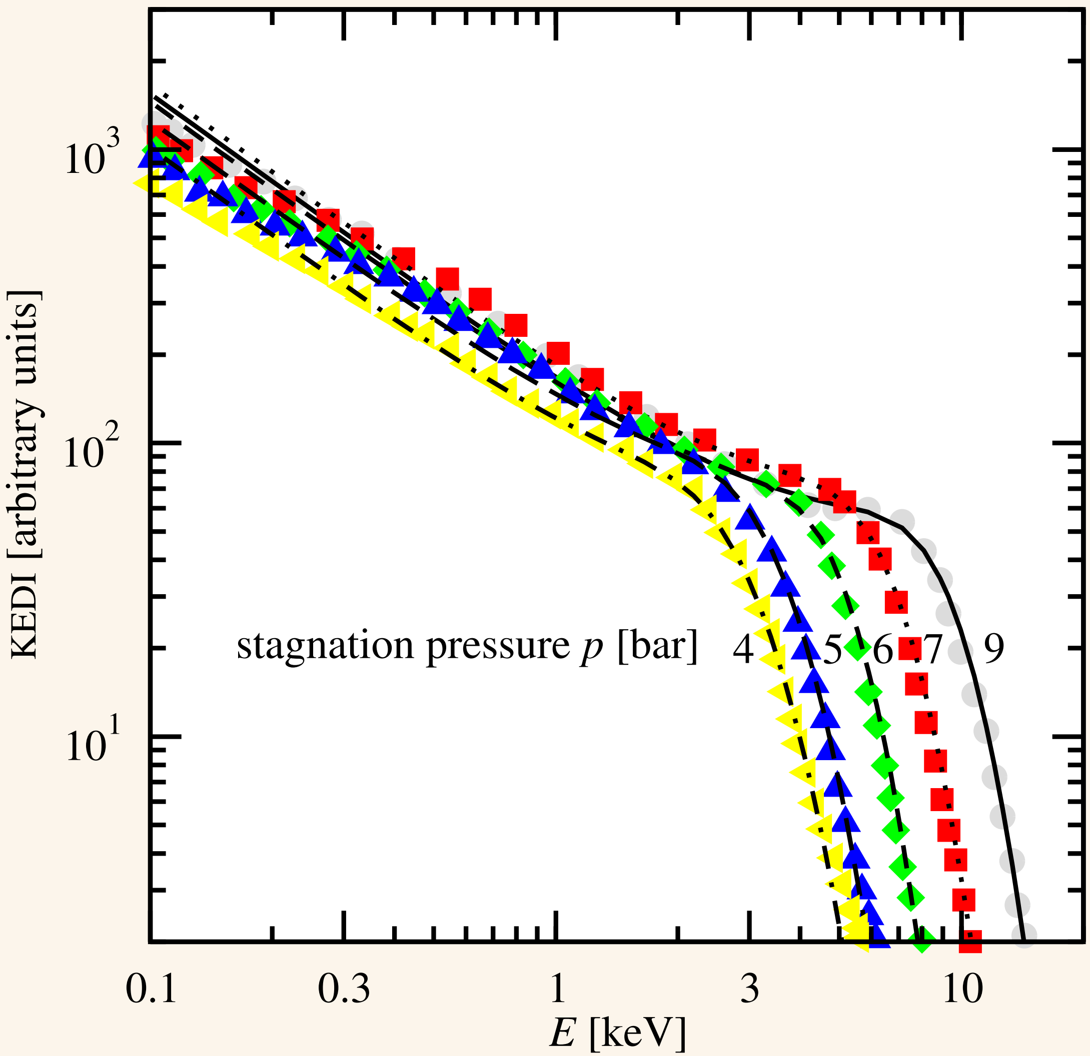
because of finite number of electrons available (e. g. hydrogen: 1, N_2: 5 per atom)



$$\frac{dP_{\mathrm{sat}}(\eta)}{d\varepsilon} = \frac{dP_{\mathrm{both}}}{d\varepsilon} - \ln\eta \, \frac{dP_{\mathrm{size}}}{d\varepsilon}$$

kinetic energy distribution of ions

model vs. experimental data for $(N_2)_N$ clusters of different sizes N
[Krishnamurthy et al. Phys. Rev. A 69 (2004) 033202]



kinetic energy distribution of ions

model vs. experimental data for $(\mathrm{N}_2)_N$ clusters of different sizes N
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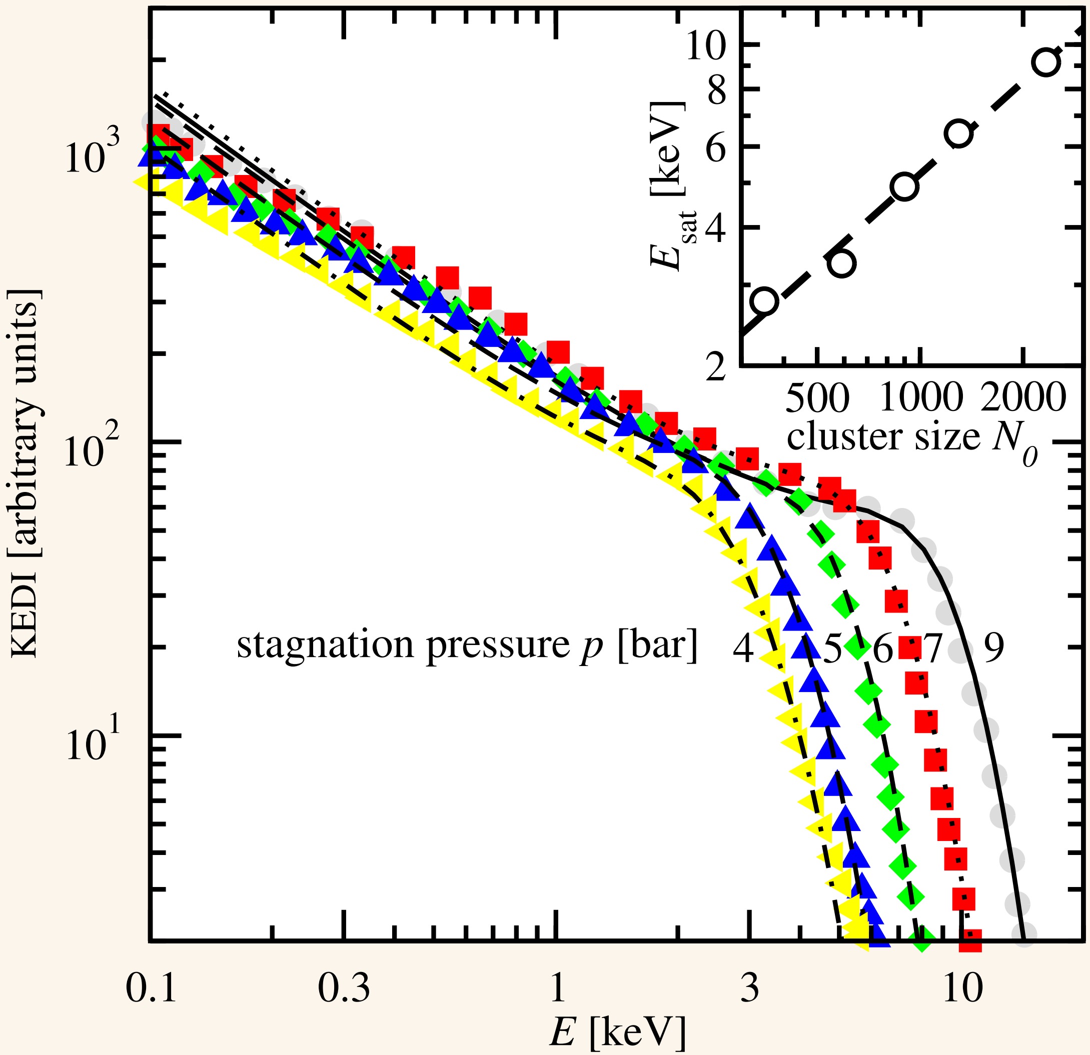
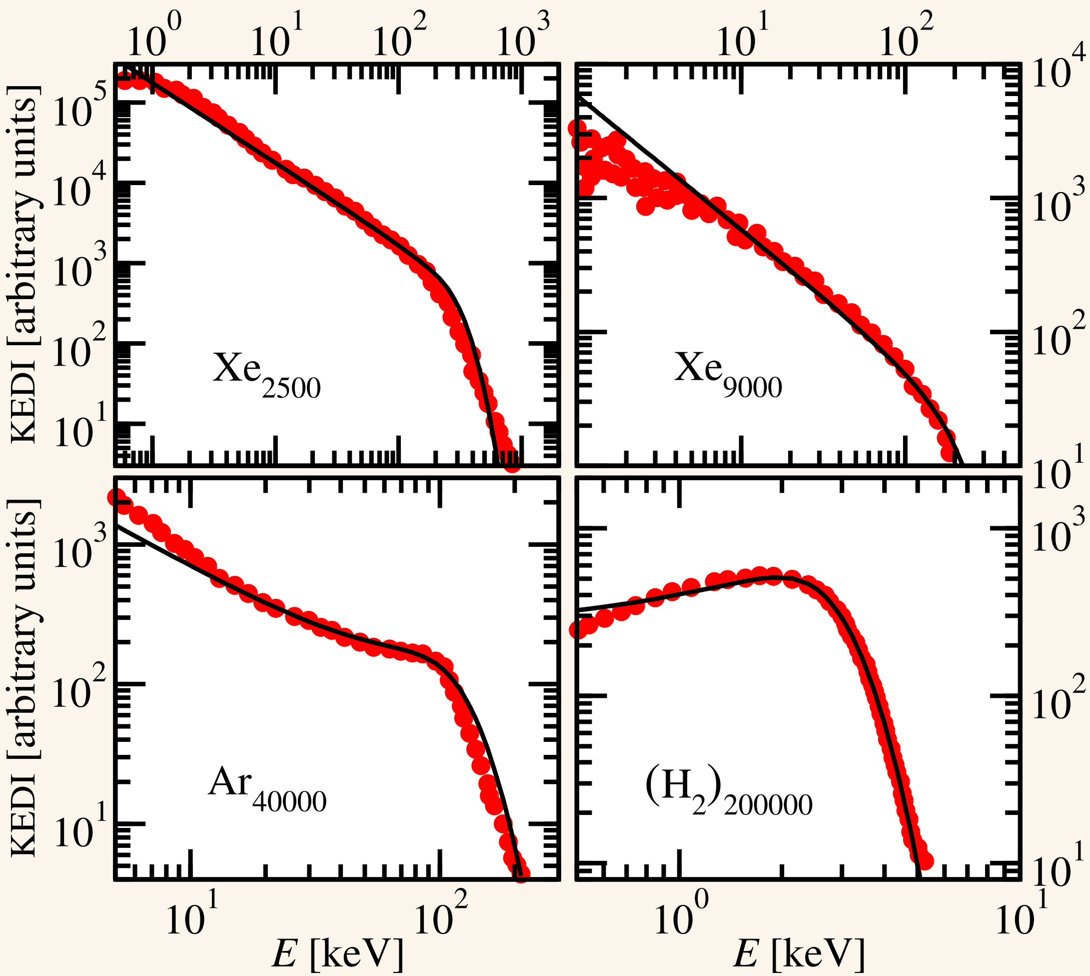


if common saturation charge for all cluster sizes

$$E_{\rm sat} \propto N_0/R \propto {N_0}^{2/3}$$

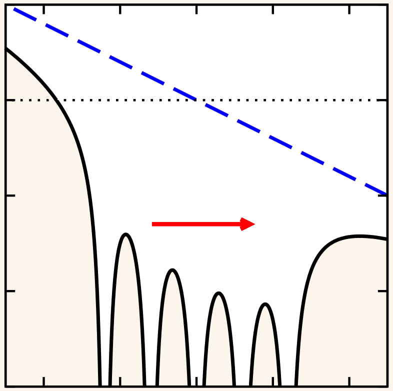
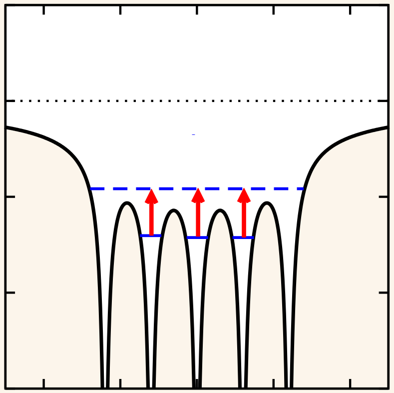
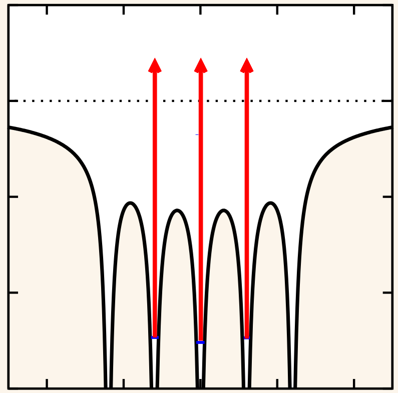
kinetic energy distribution of ions

experimental data from different groups studying different targets



ionization at shorter wavelengths / higher frequencies

[Topical Review: US, Ch Siedschlag & JM Rost, J. Phys. B 39 (2006) R 39]

	780 nm, 1.5 eV	90 nm, 13 eV	3 nm, 350 eV
Keldysh parameter	$\gamma \ll 1$	$\gamma \sim 1$	$\gamma \gg 1$
quiver amplitude	$\sim$ cluster size	$\sim$ atom size	~ 0
			
inner ionization	barrier suppression	single-photon absorption (into cluster)	single-photon absorption or auto-ionization
outer ionization	collective heating	inverse bremsstrahlung	(into continuum)

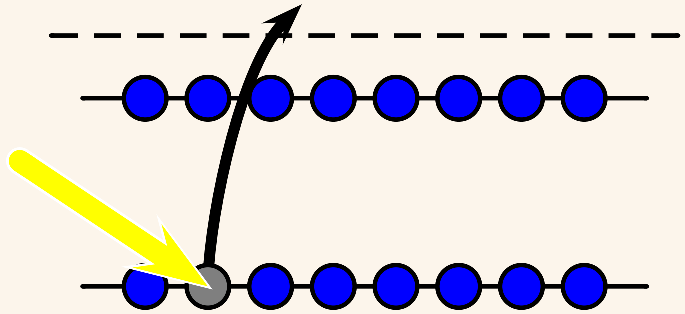
treatment of laser-cluster interaction

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

mixed quantenmechanical–classical approach

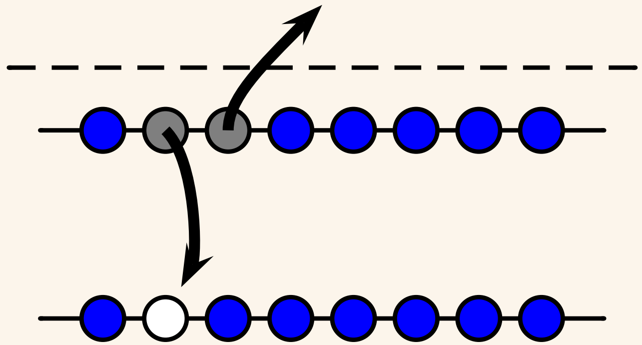
- atomic ionization and intra-atomic decays
  - statistical description by means of **quantenmechanical** transition rates (photo-ionization, auto-ionization)
- dynamics of free electrons and ions
  - propagation of **classical** equations of motion

X-FEL: ionization of atoms “inside–out”



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

$$\Gamma = I[\mathrm{au}] \cdot 0.1\,\mathrm{fs}^{-1}$$



(2) decay cascades:

— **Auger decays** $\Gamma_{\mathrm{Argon}} = 0.2 \ldots 5\,\mathrm{fs}^{-1}$

— radiative transitions

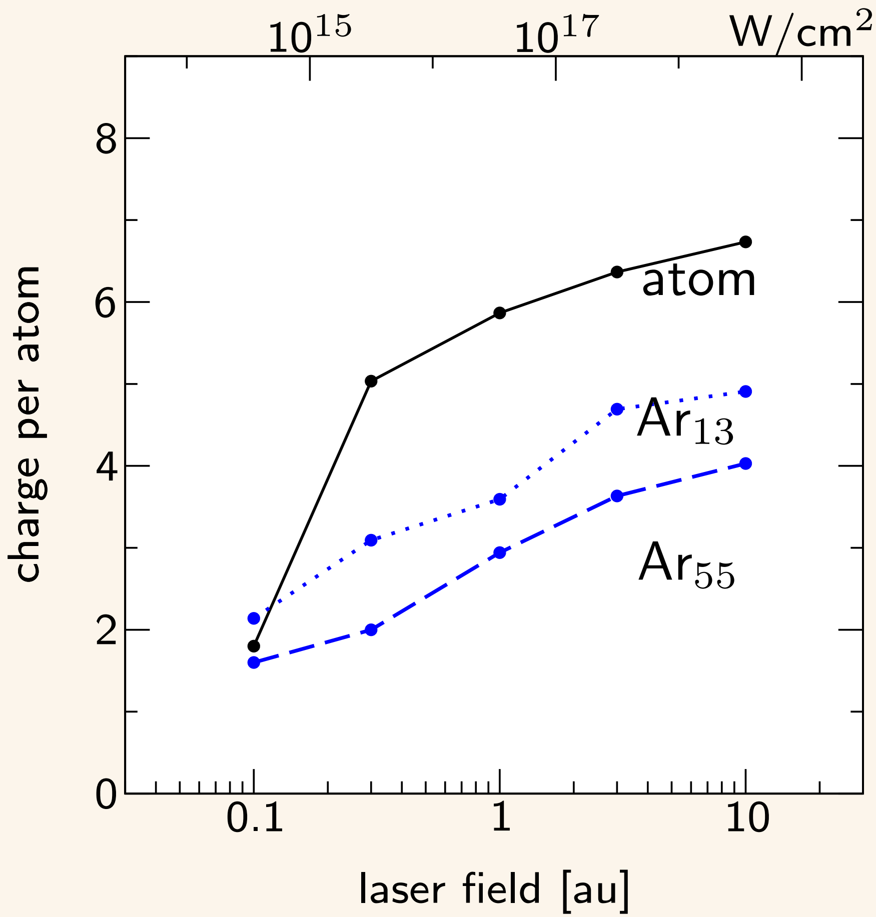
— “shake-off” processes

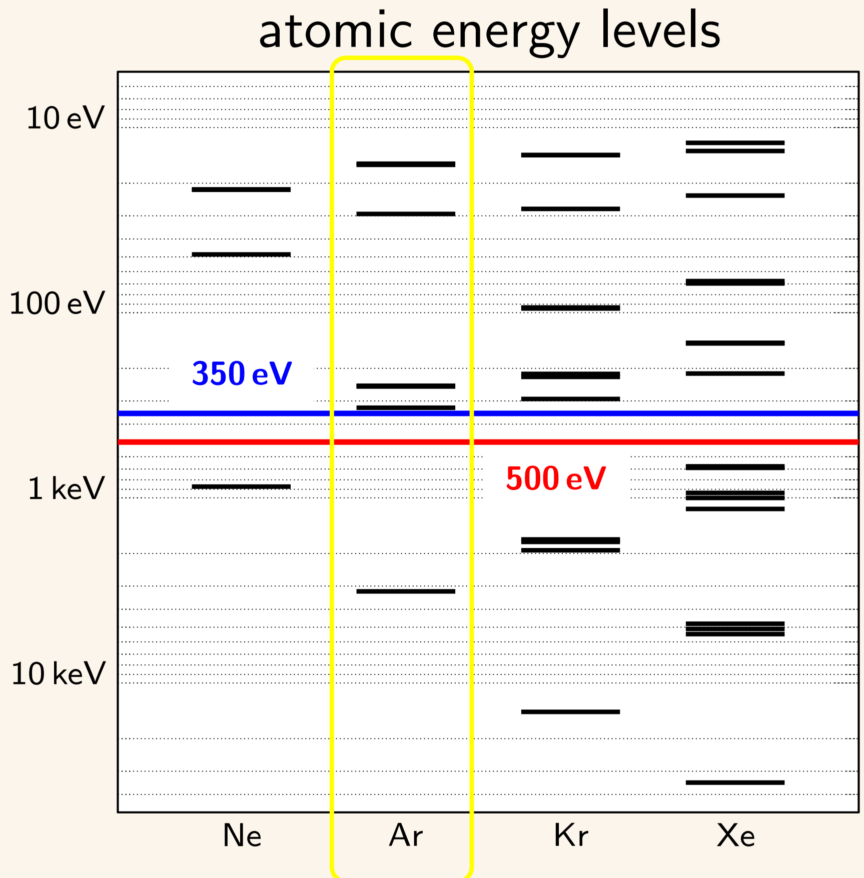
expectation: multiple photo-ionization } during pulse

fast decay cascades }

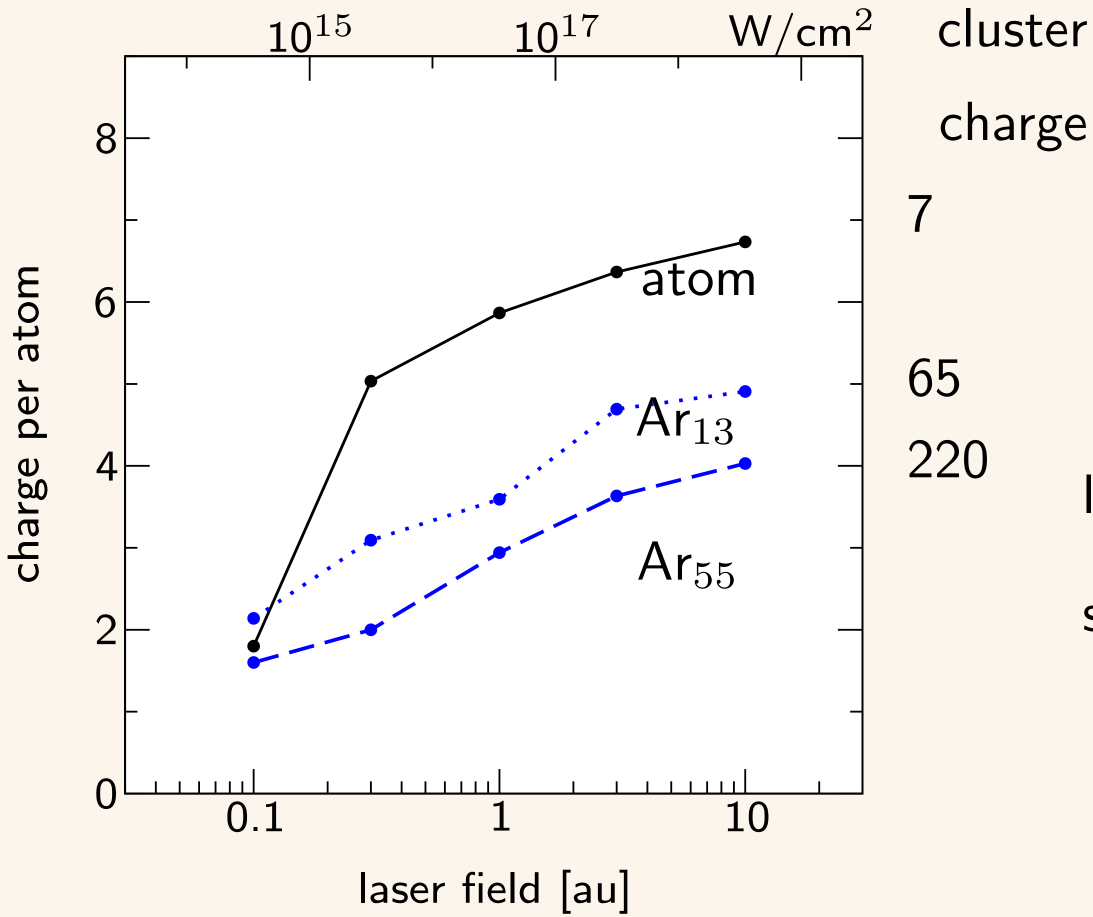
→ **enormous energy absorption**

argon cluster @ X-FEL pulse (350 eV, 80 fs)





argon cluster @ X-FEL pulse (350 eV, 80 fs)

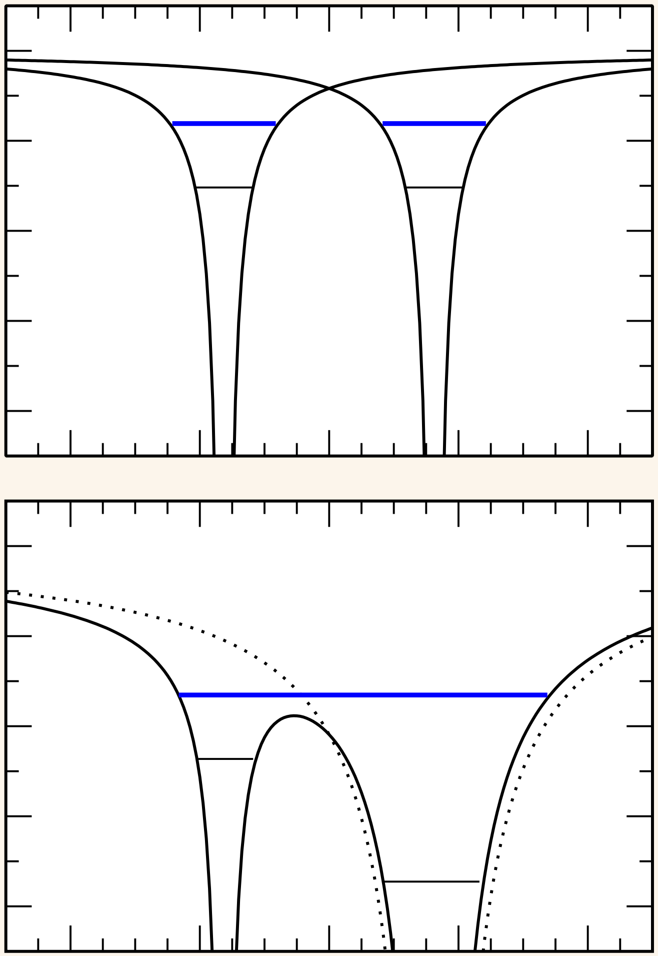


large space charge &

small quiver amplitude $\sim \dfrac{\sqrt{I}}{\omega^2}$

delocalization of valence states

delocalization of valence states



coordinate [au]

atomic photo-ionization

JMR, J. Phys. B 28 (1995) L 601

$$\Gamma \propto \left|\Psi(r_\omega)\right|^2 \qquad \text{with} \quad r_\omega = \frac{1}{\sqrt{\omega}} = 0.25\,\mathrm{a}_0$$

values close to nucleus reduced

auto-ionization

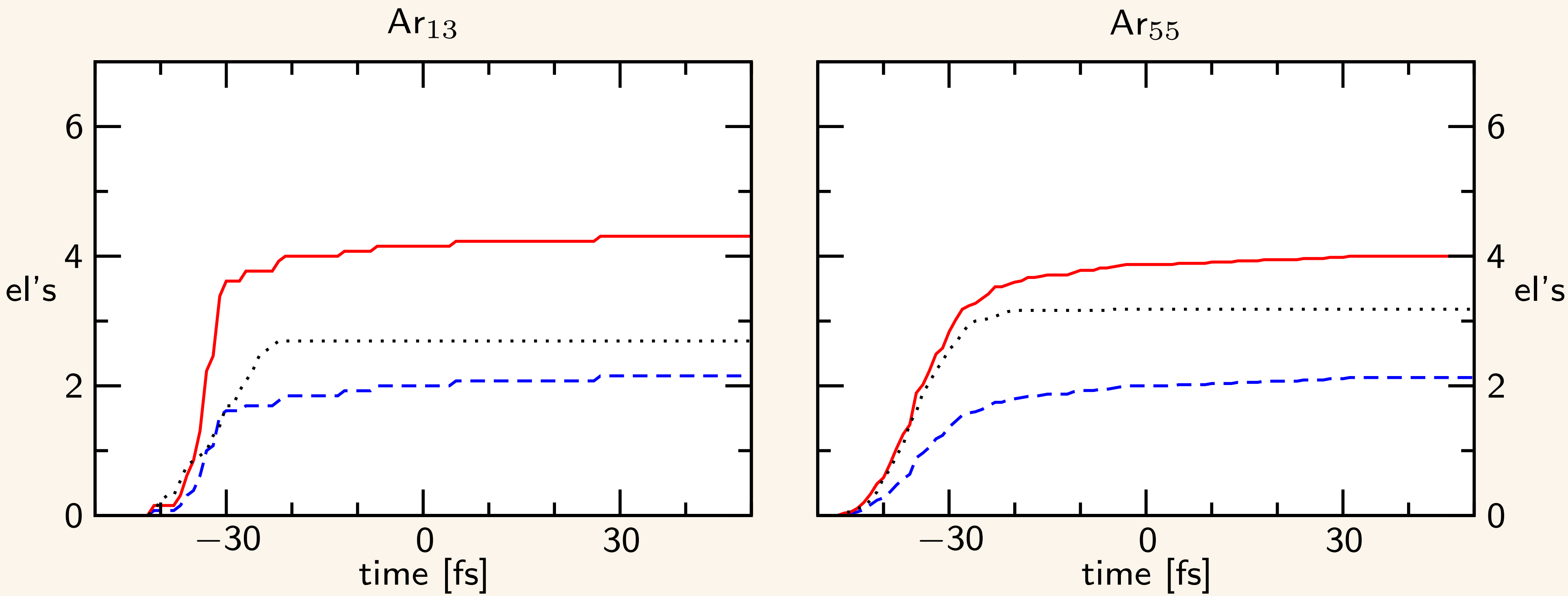
Fermi's Golden Rule

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

overlap of excited electrons and hole reduced

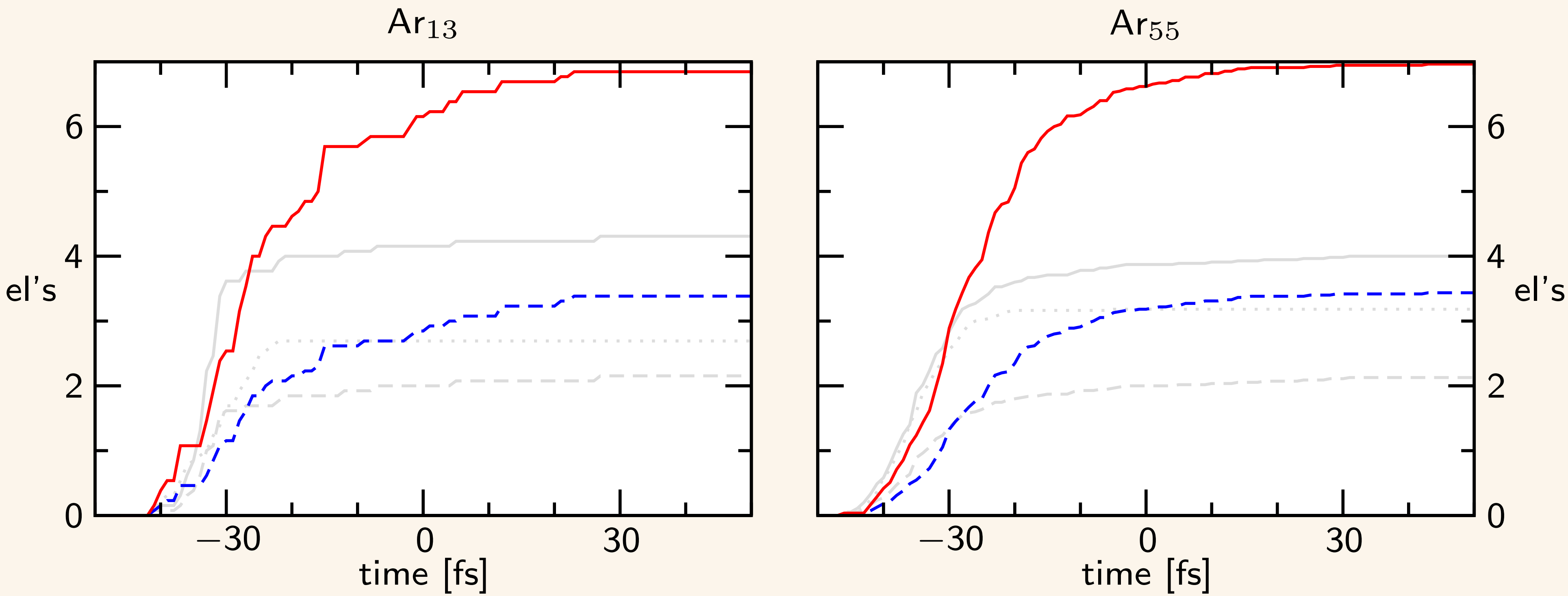
pulse (350 eV, 1 au): ionized electrons

photo-ionization tunneling photo- and auto-ionization



pulse (350 eV, 1 au): ionized electrons without tunneling

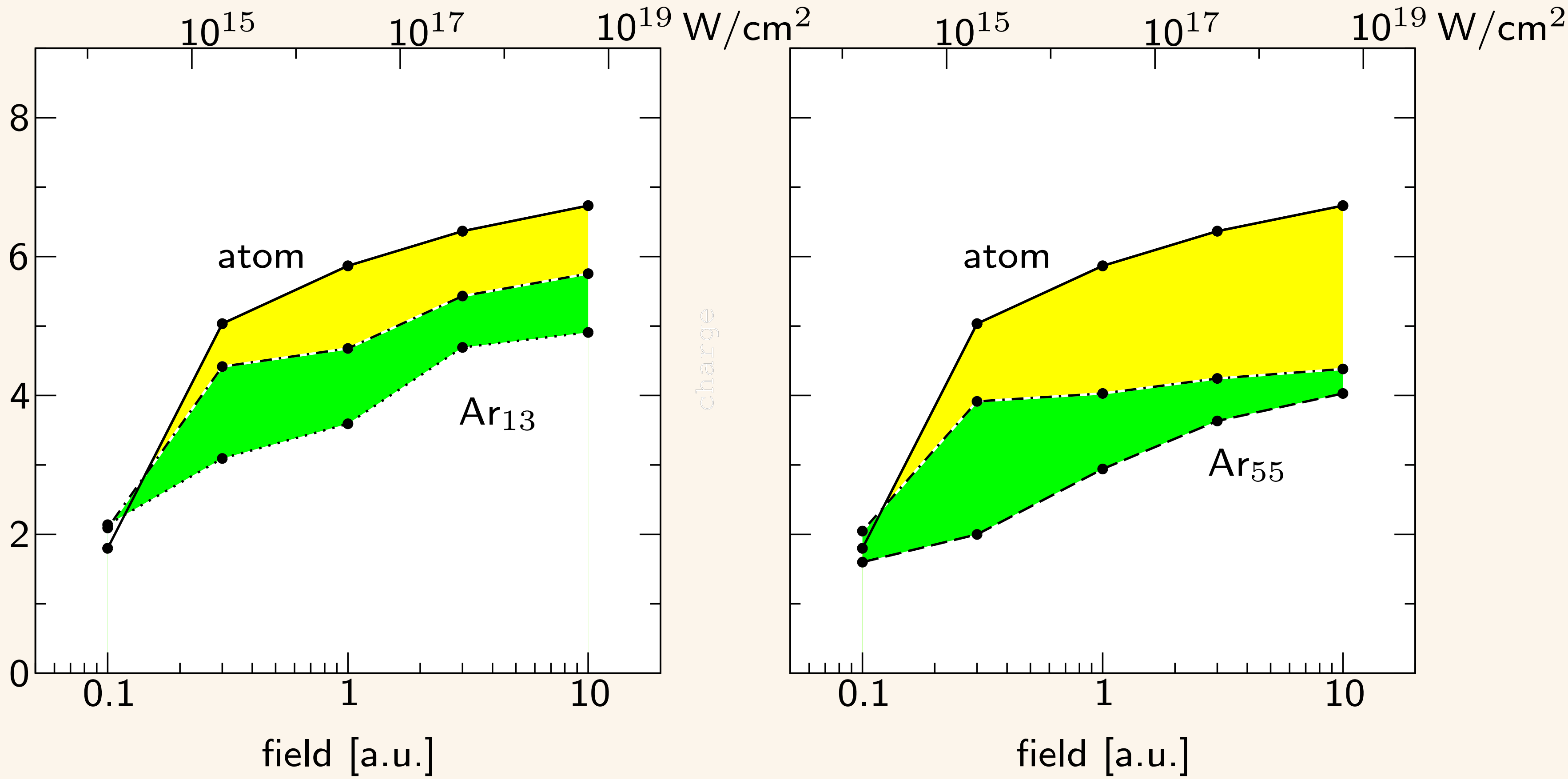
photo-ionization ~~tunneling~~ photo- and auto-ionization



clusters in X-ray laser pulses

mpipks

space-charge effect vs. delocalization effect [US & Rost, PRL 89 (2002) 143401]



The End !