

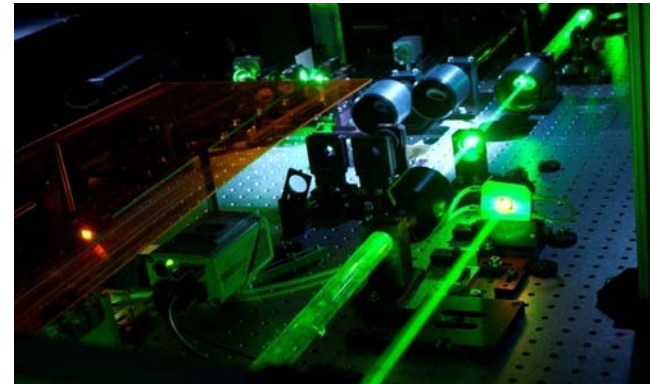
Generation of keV photons and electrons during laser-cluster interaction



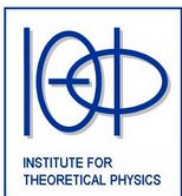
***C. RAMOND, J.HABIB, E. LAMOUR, C. PRIGENT,
R. REUSCHL, J-P. ROZET, M. TRASSINELLI, D. VERNHET***



O. GOBERT M. PERDRIX



Collaborations:



C. DEISS, J. BURGDÖRFER
TU-Wien – Institute for Theoretical
Physics, Austria



G. SCHIWIETZ

Outline

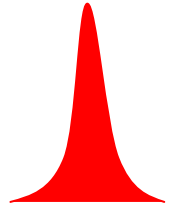
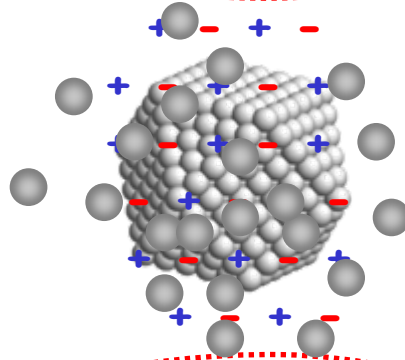
- some properties of intense laser - cluster interaction
correlation between X-ray emission and energetic electrons?
- Interaction dynamics:
mechanisms responsible for the X-ray production
- results focused on the last experiments performed:
evidence of an ignition process
correlation between X-ray yield and electron spectra
- conclusions & perspectives

Clusters in intense laser fields

Strong optical fields

$$F > 10^9 \text{ V/cm}$$

$$E_{\text{hv}} \sim 1.6 \text{ eV} / 3.2 \text{ eV}$$



Creation of nanoplasmas

time scale \sim pulse duration



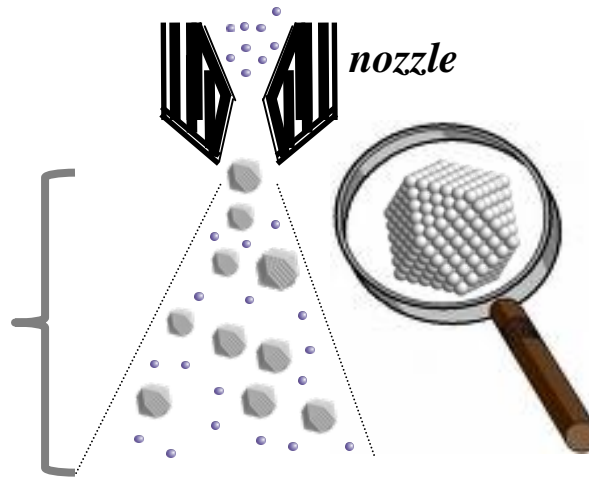
**Expansion and explosion of
heated clusters**

Van der Waals clusters...why?

Finite systems with variable sizes : $\tilde{N} = 10^3 - 10^6$ at/cl ($\phi \sim 1$ to 30 nm)

$\lambda_{laser} \gg \text{skin depth} \geq \phi_{cluster} \Rightarrow \text{uniform field inside each cluster}$

Cluster jet



□ **low mean atomic density**

$\sim 10^{14} - 10^{17}$ at/cm³

well separated ($\sim 1-10$ μm)

no problem of laser propagation

no x-ray absorption

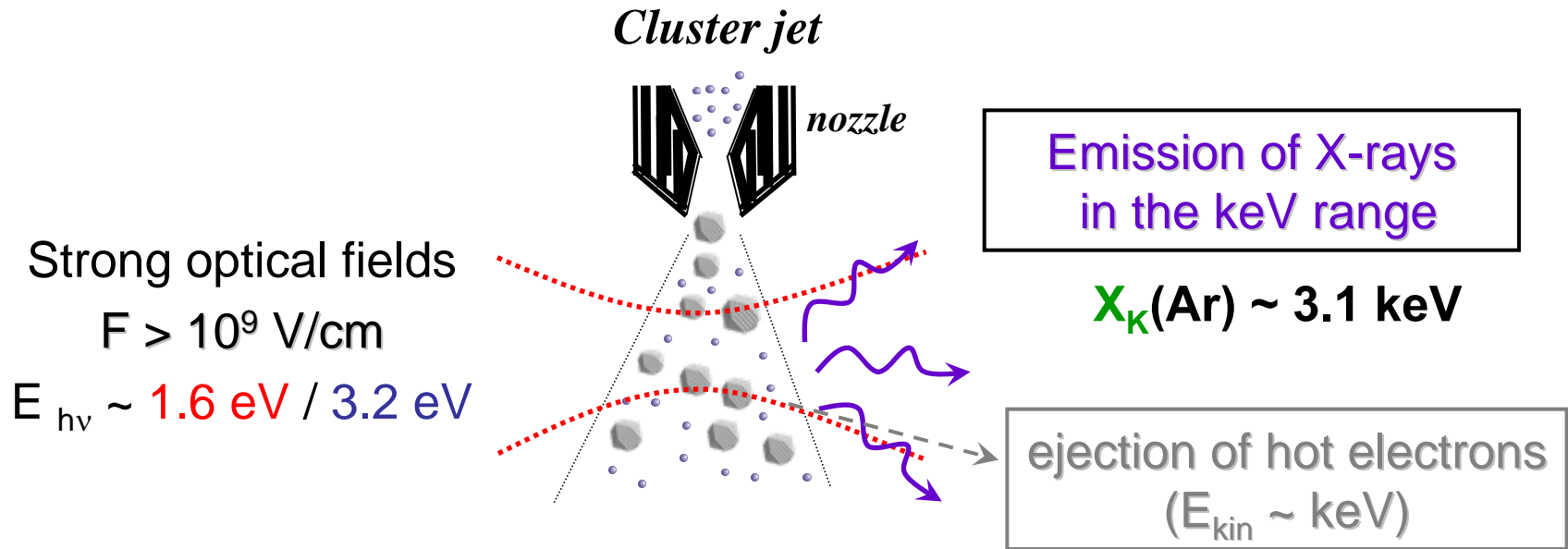
□ **high local density**

close to solid $\sim 10^{22}$ at/cm³

enhanced energy coupling
between light & matter

A cluster jet combines advantages of gaseous and solid targets

Van der Waals clusters in intense laser fields



**Direct insight into the early evolution of the nanoplasma
(time-scale comparable to the laser pulse duration)**

But never under the same experimental conditions!!!

An incomplete list of several studies:

H. M. Milchberg *et al* PRE **62** (2000)

J.P. Rozet *et al* Phys.Scripta **T92** (2001)

V. Kumarappan *et al* PRA **63** (2001)

F. Dorchies *et al* PRE **71** (2005)

L.M. Chen *et al* PRL **104** (2010)

Y.L. Shao *et al* PRL **16** (1996)

L.M. Chen *et al* PRE **66** (2002)

E. Springate *et al* PRA **68** (2003)

V. Kumarappan *et al* PRA **67** (2003)

Y. Liu *et al* Phys. of Plasma **16** (2009)

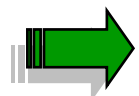
Van der Waals clusters in intense laser fields

What we understand about the X-ray production

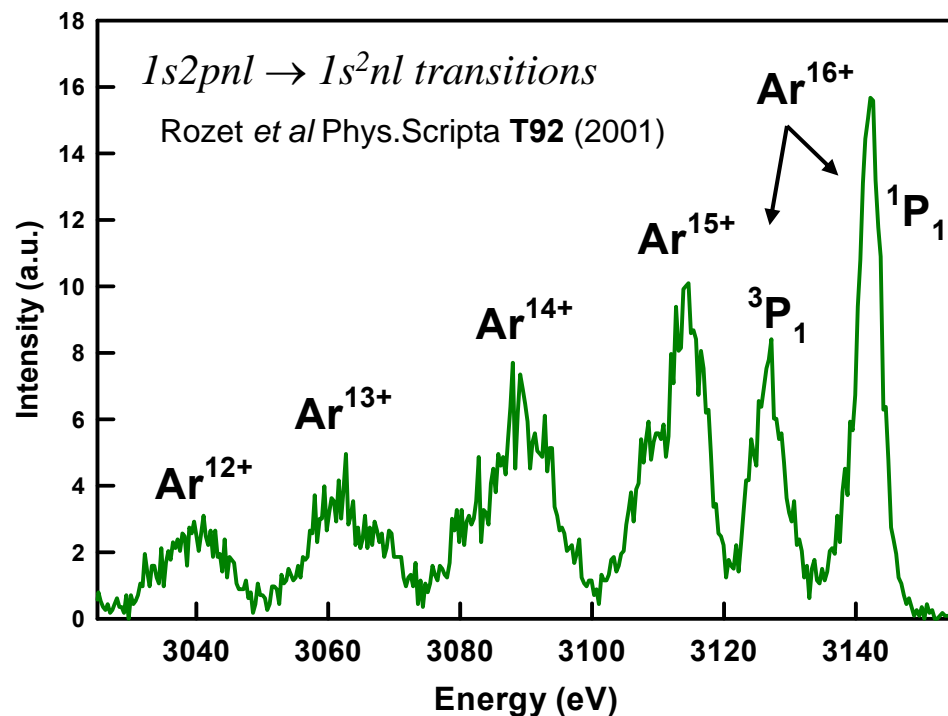
Emission of X-rays
in the keV range



Inner shell vacancy
production



Deexcitation of
HCI with inner shell
vacancies



up to Ar¹⁶⁺ with $\tau(1P_1) = 15$ fs

time scale down to some fs

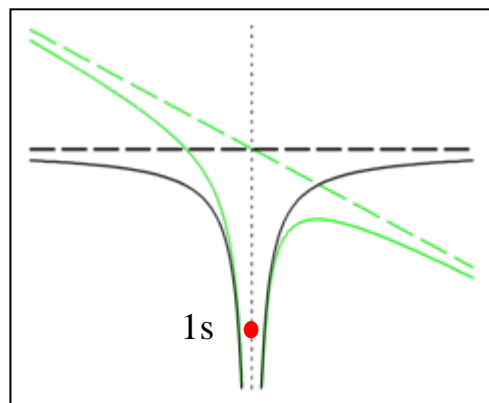
Van der Waals clusters in intense laser fields

What we understand about the X-ray production

Emission of X-rays
in the keV range

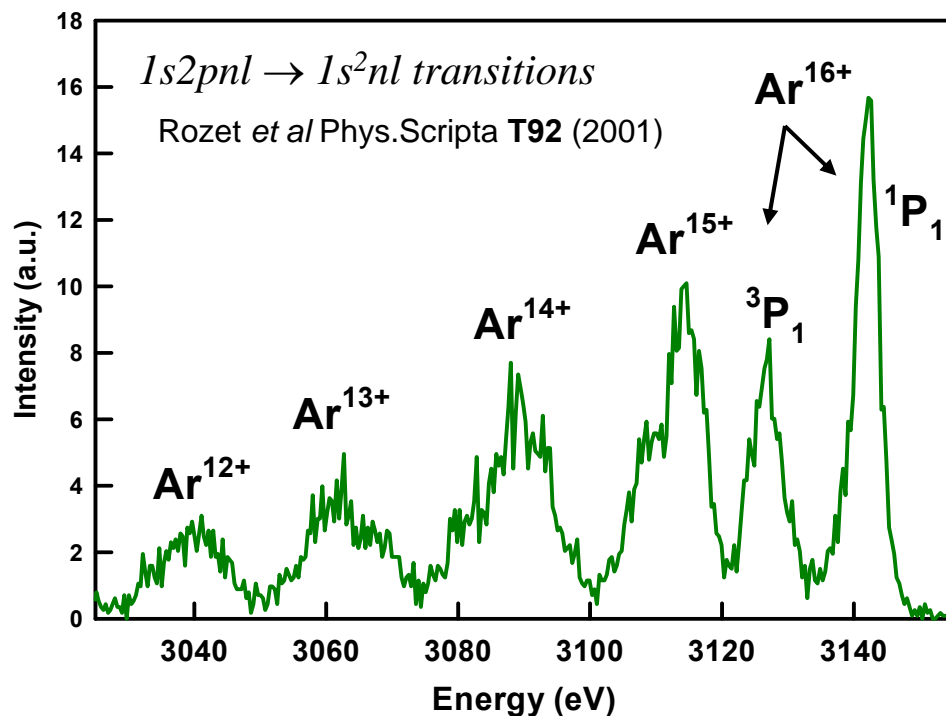


Inner shell vacancy
production



Inner shell vacancy by OFI

$\text{Ar}^{16+} (1s) \rightarrow 4 \times 10^{21} \text{ W/cm}^2$



up to Ar¹⁶⁺ with $\tau(^1P_1) = 15$ fs

time scale down to some fs

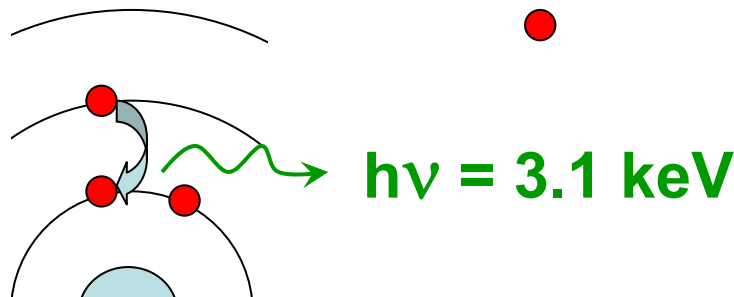
Van der Waals clusters in intense laser fields

What we understand about the X-ray production

Emission of X-rays
in the keV range

Inner shell vacancy
production

Electron impact
ionization



Fast electrons
with $E = E_K \sim 3 - 4 \text{ keV}$
for argon ions

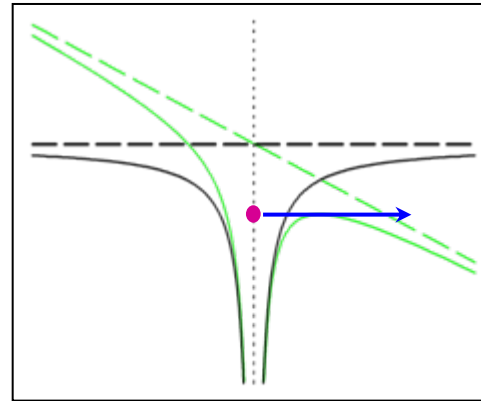
Understanding of the **electron heating** mechanisms
and the link between energetic electrons and X-ray

Outline

- some properties of intense laser - cluster interaction
correlation between X-ray emission and energetic electrons?
- Interaction dynamics:
mechanisms responsible for the X-ray production
- results focused on the last experiments performed:
evidence of an ignition process
correlation between X-ray yield and electron spectra
- conclusions & perspectives

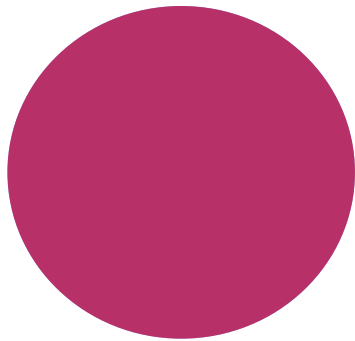
Scenario for electron heating mechanisms in the MF CTMC

Developed by the group of J.Burgodörfer



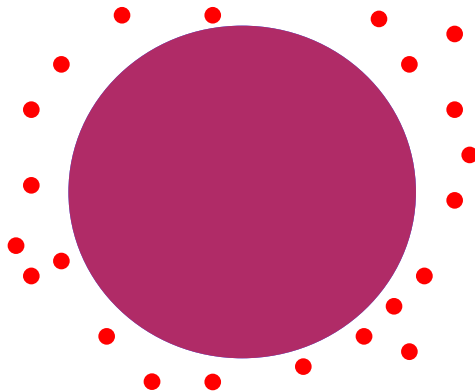
Optical field ionization

→ a cold nanoplasma



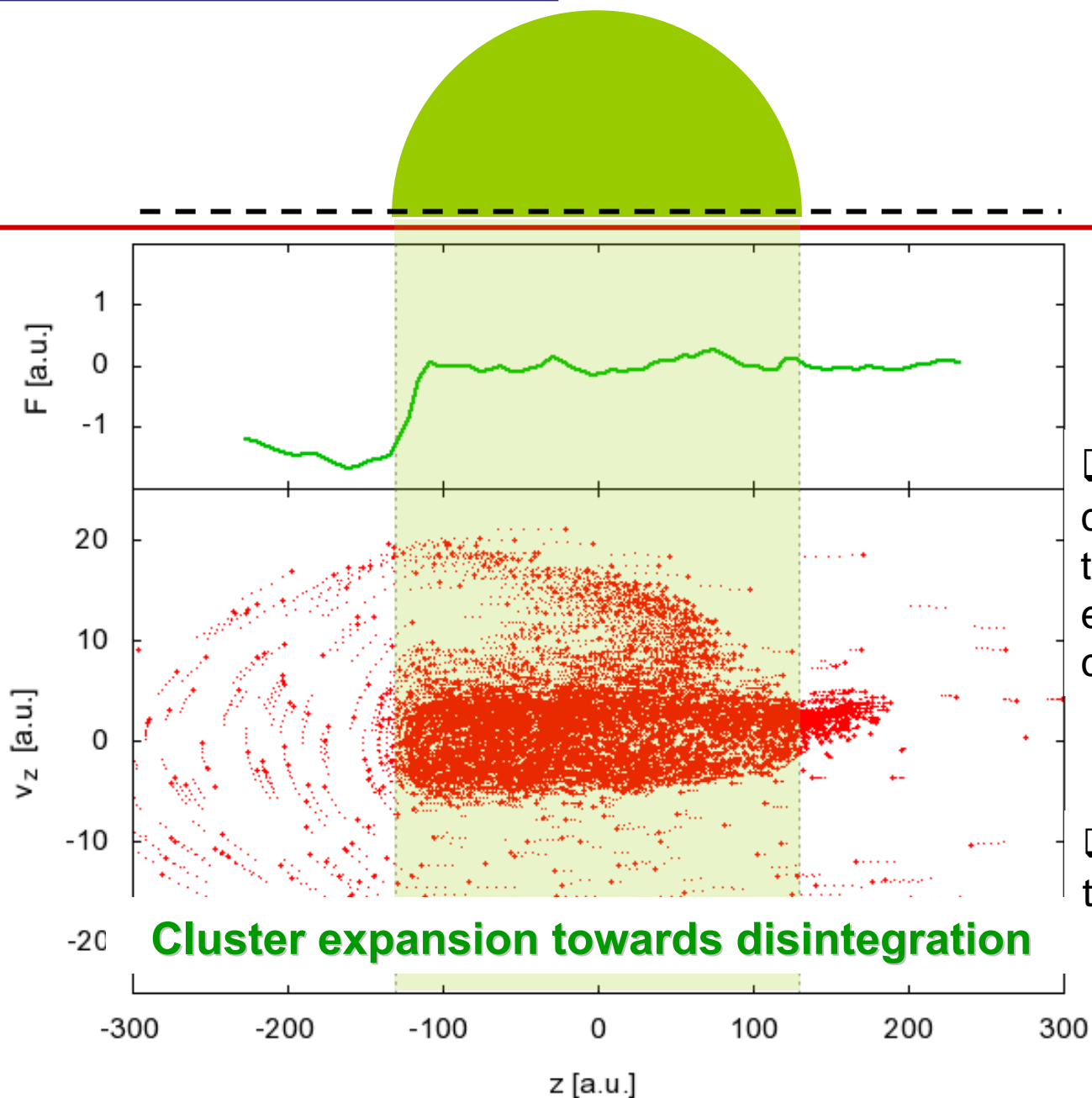
Polarizable sphere

- screening of the laser field inside the cluster
- but on the poles electric field enhanced



Outer ionization : electrons leave the cluster

Build-up of a positive charge on the cluster surface



Cluster expansion towards disintegration

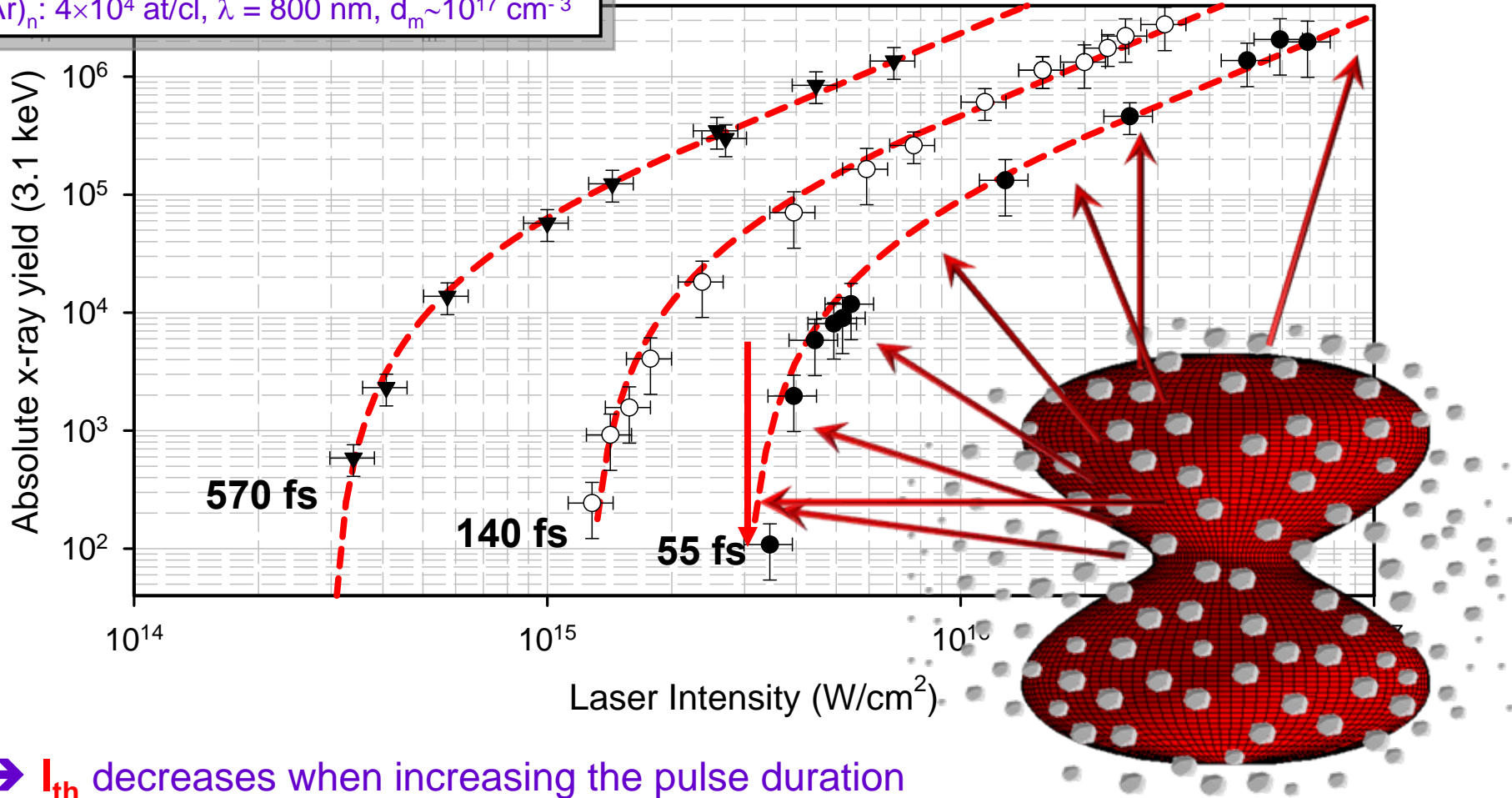
□ a fraction of electrons can gain energy due to the presence of strong electric fields at the cluster poles

□ electrons accelerated through back the cluster
→ K-shell ionization

Absolute X-ray yields vs laser intensity for different τ

(Ar)_n: 4×10^4 at/cl, $\lambda = 800$ nm, $d_m \sim 10^{17}$ cm⁻³

$$N_X \propto P_K(I_{\text{peak}}, \tau) \times V_{\text{eff. foc.}}(I_{\text{peak}}, \tau)$$

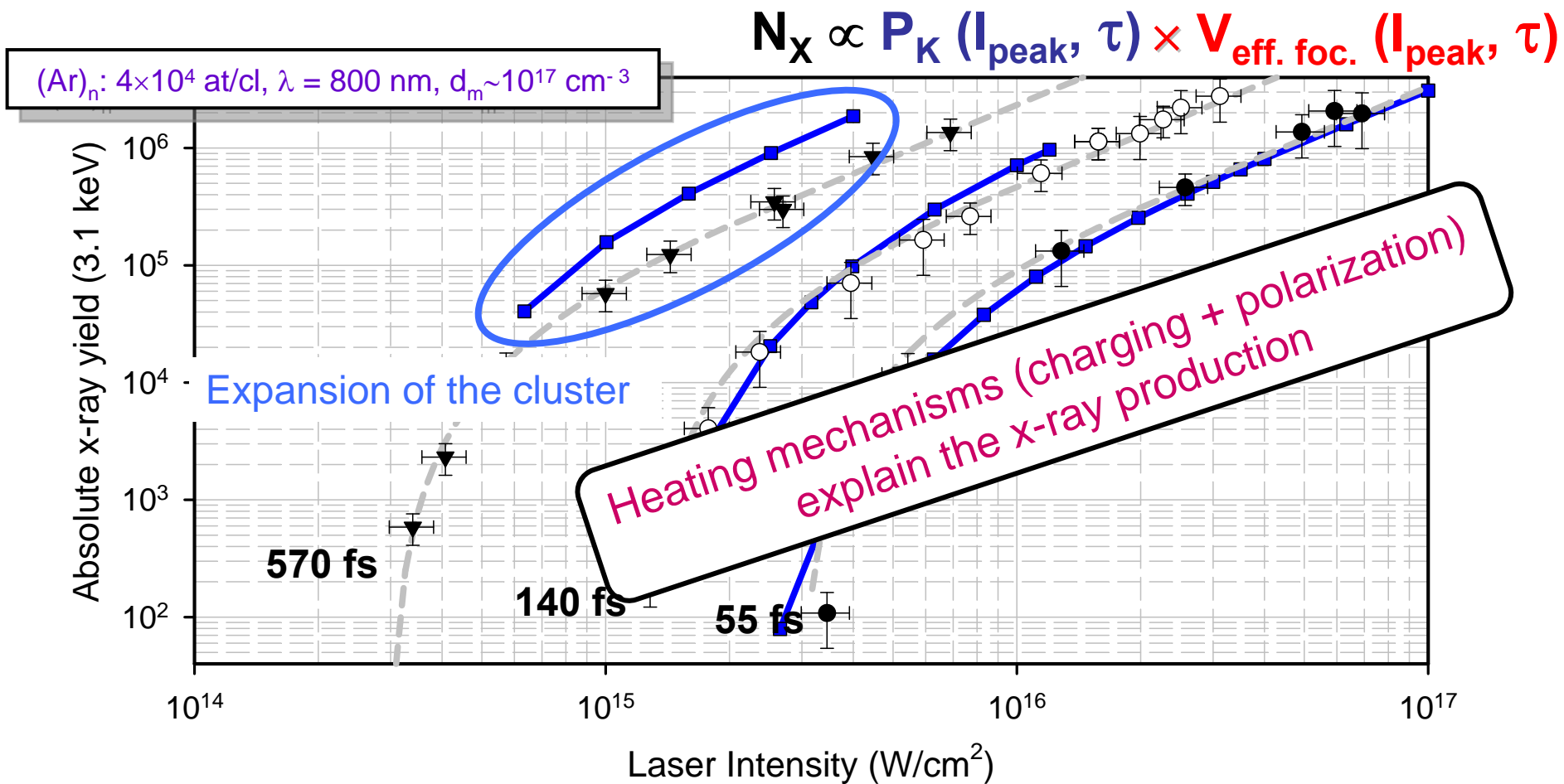


→ I_{th} decreases when increasing the pulse duration
down to an unexpected low value

→ X-ray yields well reproduced by the $V_{\text{eff. Foc}} = f(I_{\text{peak}}/I_{\text{th}})$.

↑ *laser*

Comparison with CTMC simulation



- ➔ Intensity & pulse duration dependences well reproduced
- ➔ Good prediction of the intensity threshold I_{th}
- ➔ Discrepancy for long pulse duration: *role of the ion dynamics i.e. cluster expansion*

Van der Waals clusters in intense laser fields

Emission of X-rays
in the keV range

Inner shell vacancy
production

Electron impact
ionization

Scenario for the heating mechanisms in the MF CTMC

- ✓ Charging-up of the cluster
- ✓ Dynamic polarization of the cluster

➔ Electrons are heated to at least 3-4keV

Our goal: improve the precision on I_{th}

Energy distribution of the electrons & link it to the X-ray emission

- ***Quantitative measurements of***
 - ✓ absolute photon emission yields
 - ✓ high energy electron distribution

as a function of different parameters

- ✓ I_{laser} , τ , polarization....

Outline

- some properties of intense laser - cluster interaction
correlation between X-ray emission and energetic electrons?
- Interaction dynamics:
mechanisms responsible for the X-ray production
- results focused on the last experiments performed:
evidence of an ignition process
correlation between X-ray yield and electron spectra
- conclusions & perspectives

Experimental set-up

Charge state distribution



reproducible experimental conditions

Van Der Waals clusters

shot-by-shot

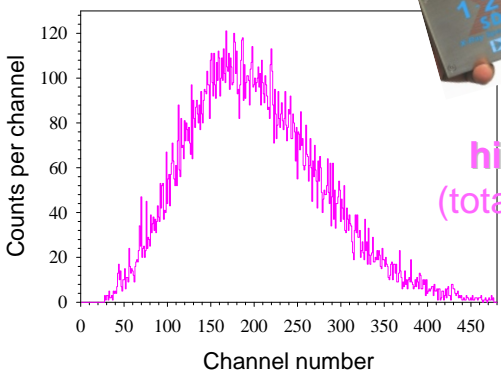
- noble gases: Ar, Kr and Xe
- nanometer size: 10 - 40 nm

10^6 at./cl

Size controlled

the backing pressure P_0

high resolution and high transmission spectrometer



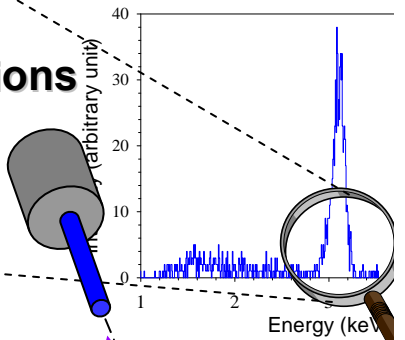
Si detector high transmission (total number of x-rays)

Cluster jet (Ar, Kr ou Xe)

Skimmer In or out

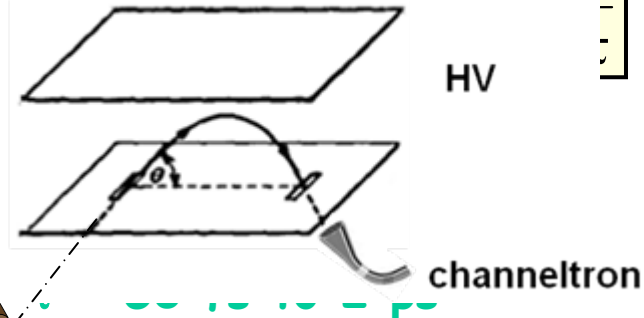
Focusing lens

LUCA Repetition rate 20 Hz

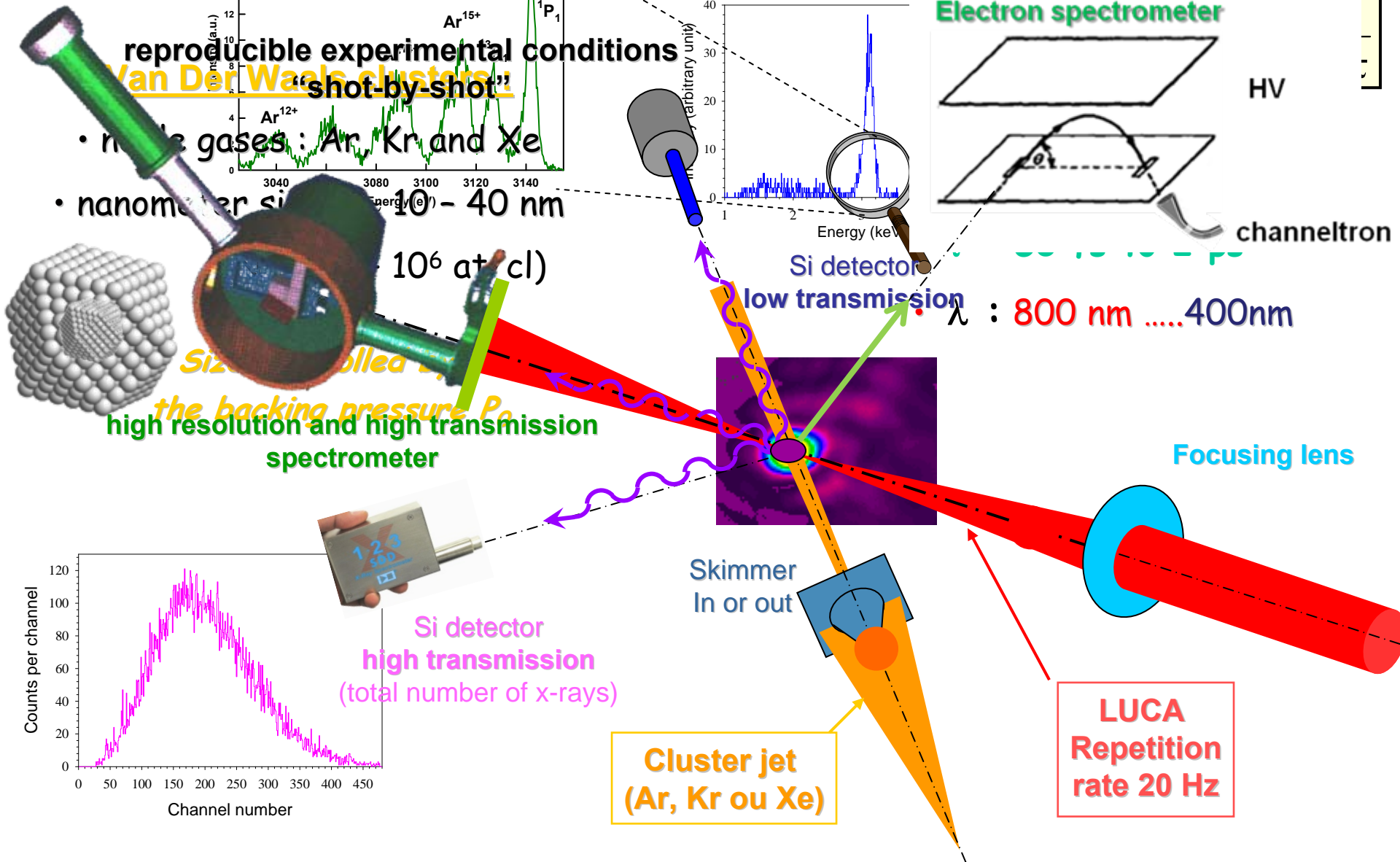


Si detector low transmission

Electron spectrometer

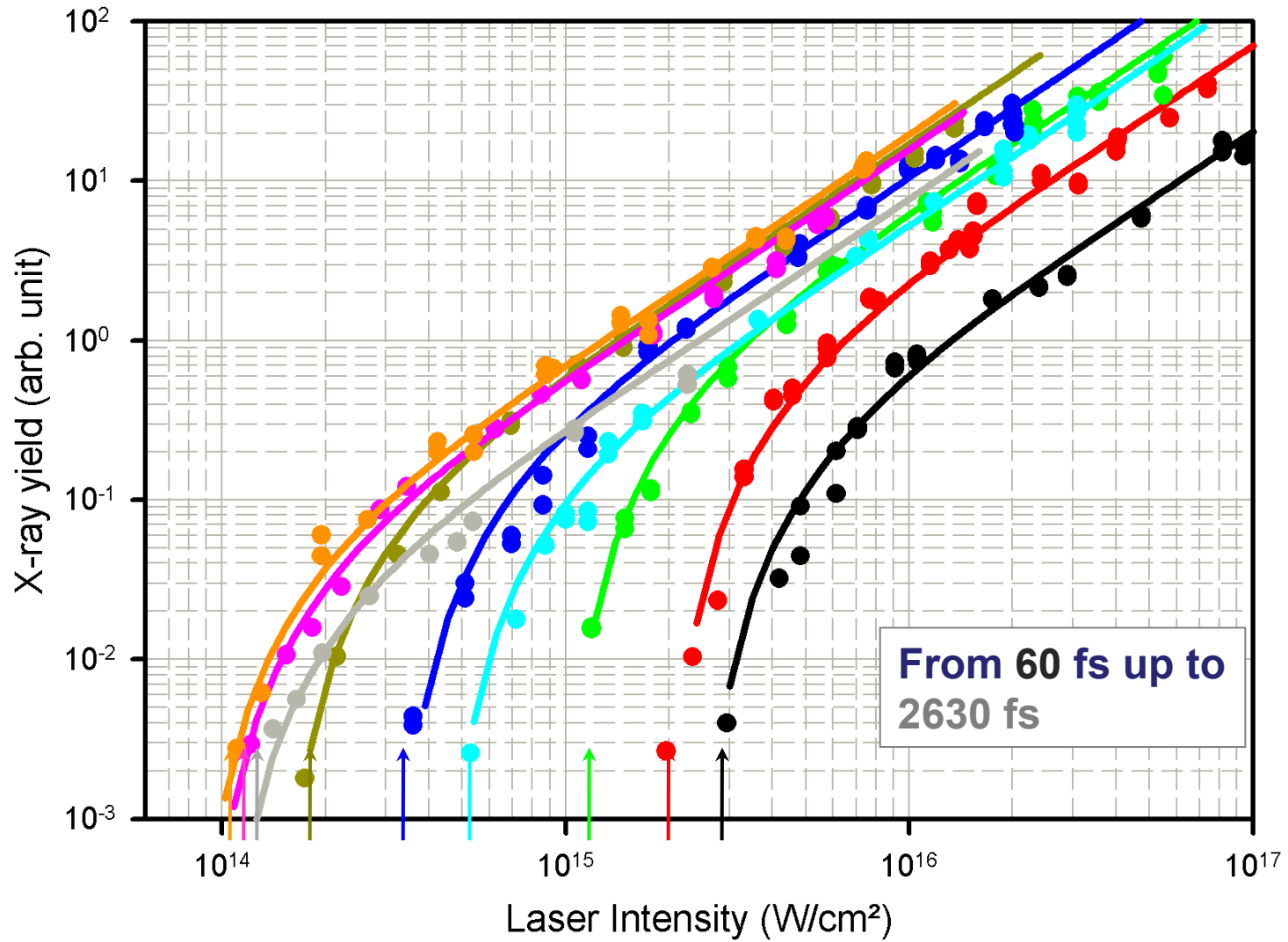


λ : 800 nm 400nm

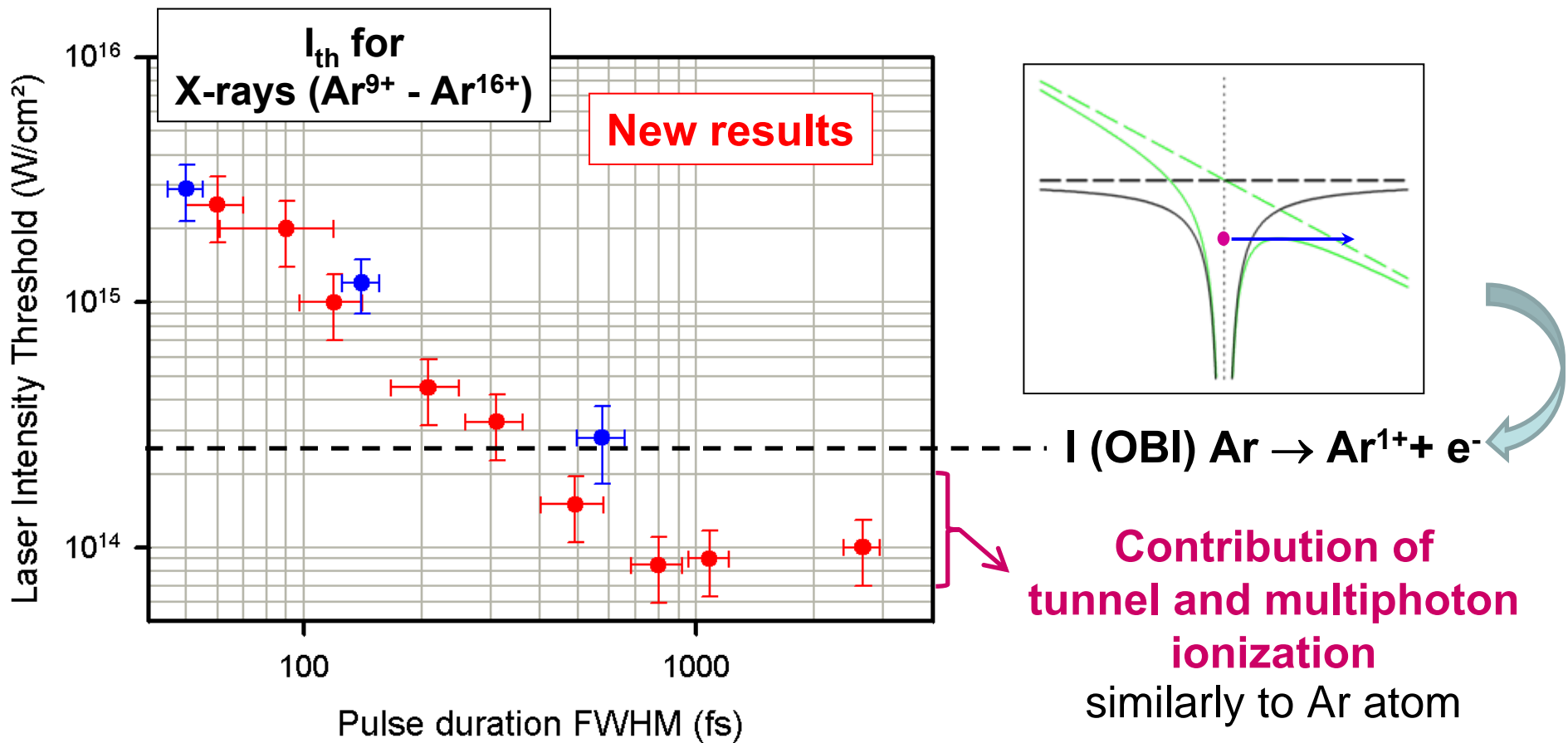


Absolute X-ray yields vs laser intensity for many pulse durations

$(\text{Ar})_n$: 2×10^5 at/cl, $\lambda = 800$ nm, $d_m \sim 10^{14}$ cm $^{-3}$; **PRELIMINARY RESULTS**



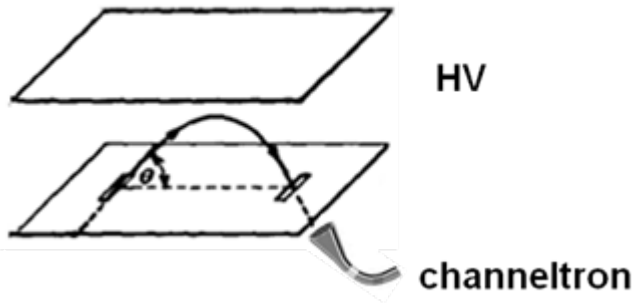
Intensity threshold for X-ray production versus pulse duration, τ



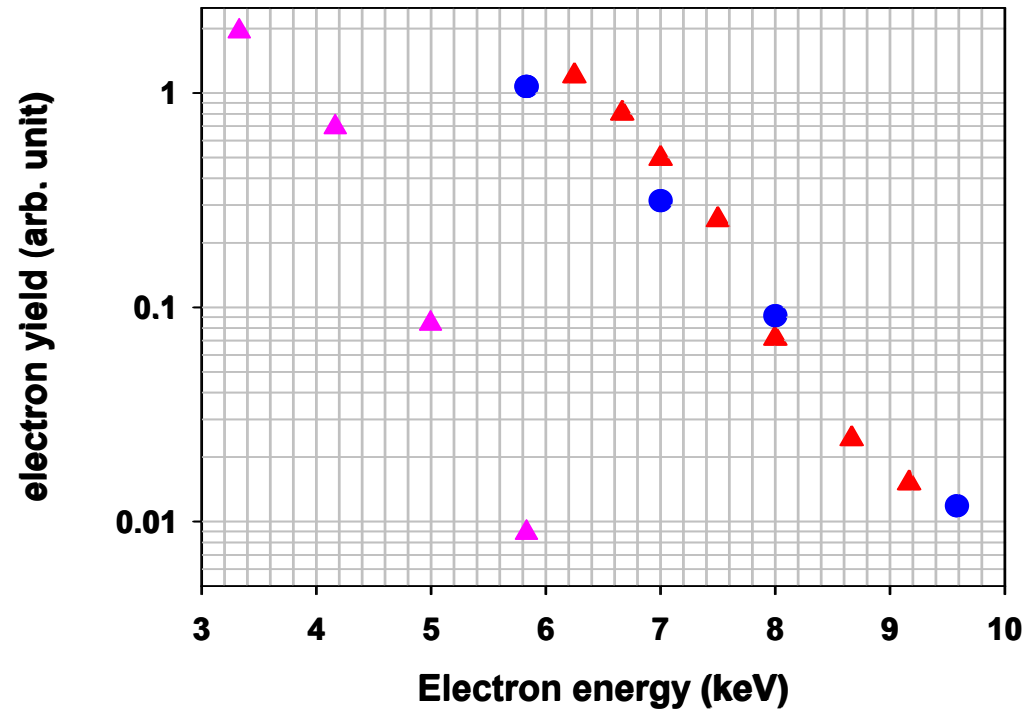
**Production of ($Ar^{1+} + e^-$) is
the ignition process for X-ray production from HCl**

Correlation between X-ray and electron yields

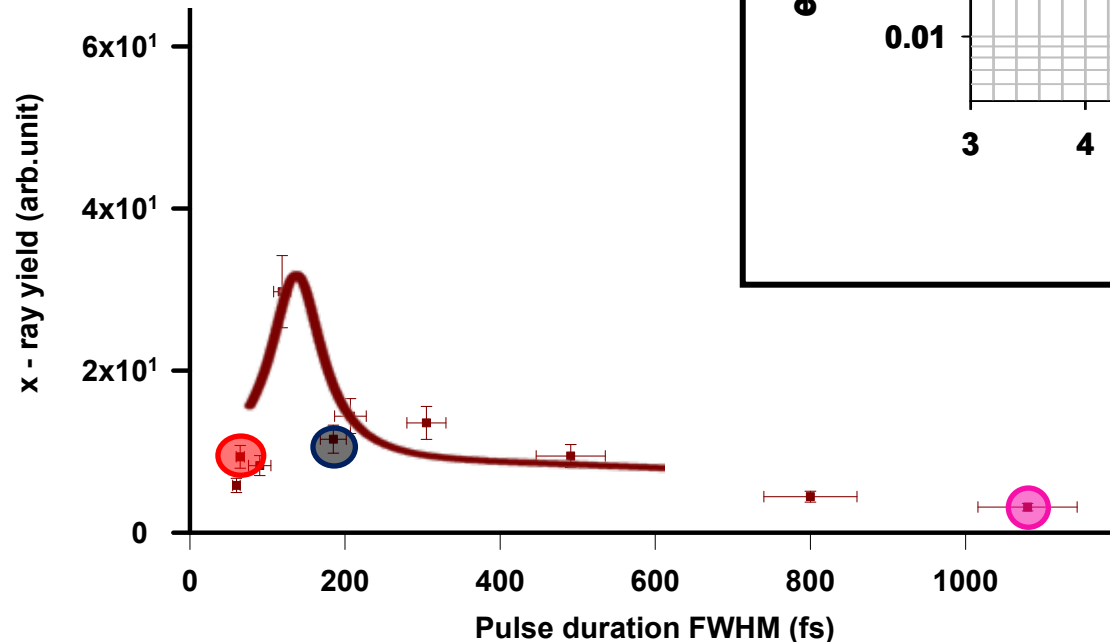
Electron spectrometer



Electron energy distribution



Preliminary results



same X-ray yield \rightarrow same $P_E(e^-)$

X-ray yield/3 \rightarrow lower $P_E(e^-)$

Conclusions & perspectives

- ✓ Absolute X-ray yield measurements under well controlled conditions provide insight of the cluster dynamics on a very short time scale
- ✓ Production of Ar^{1+} is the ignition process for X-ray production emitted from Ar^{9+} - Ar^{16+}
- ✓ Strong correlation between X-ray and high energy electron yields
... data analysis and further theoretical developments under progress
- ✓ Systematic studies have to be performed with a 400nm laser wavelength
better implementation of the expansion of the cluster in the theoretical model

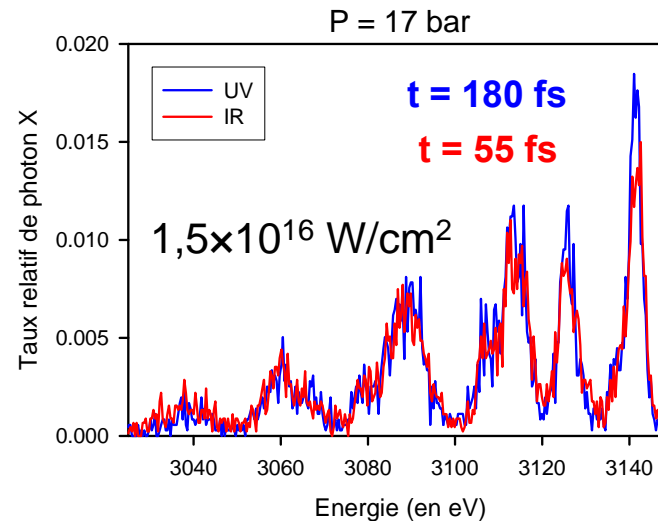
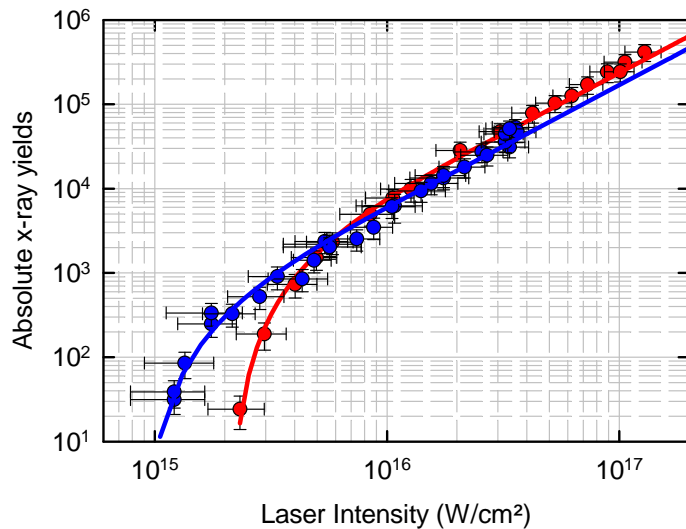
Thank you for your attention

Experiments under progress at Saclay

Doubling-frequency has been realized to obtain laser pulses @400nm

✓ Energy up to 15mJ

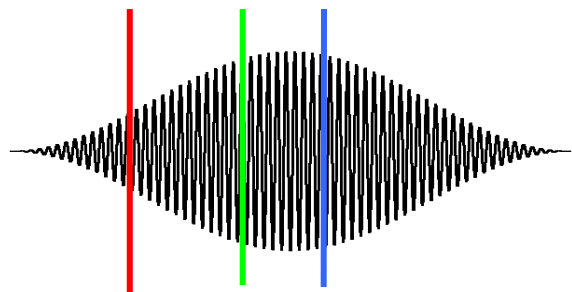
✓ Pulse duration between 60fs and 1 ps.



Influence of τ
at 400 nm

Both X-ray and electron spectroscopy are performed
→ Understand the heating mechanisms @400nm
And gain insight in the cluster expansion dynamics

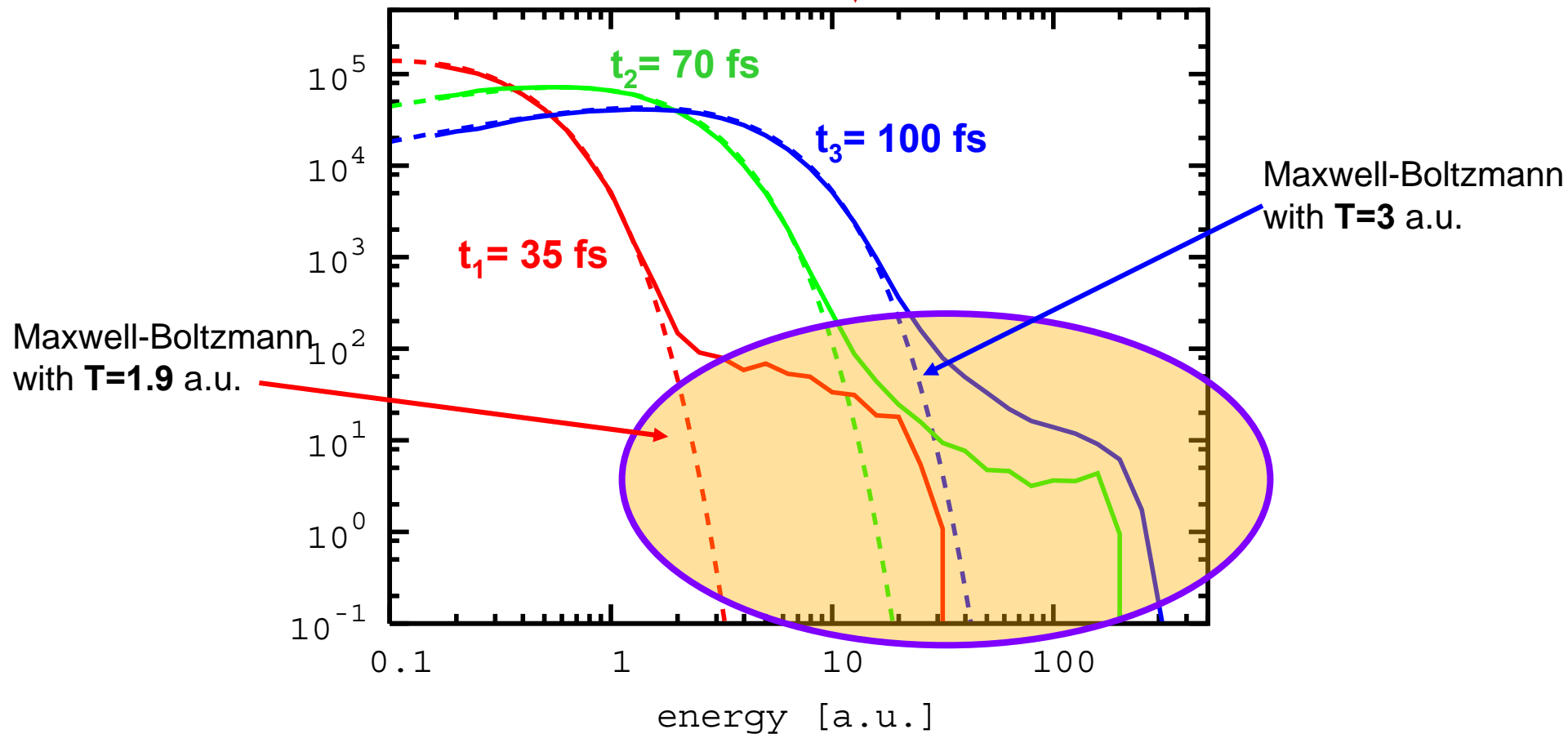
a mean field classical transport simulation

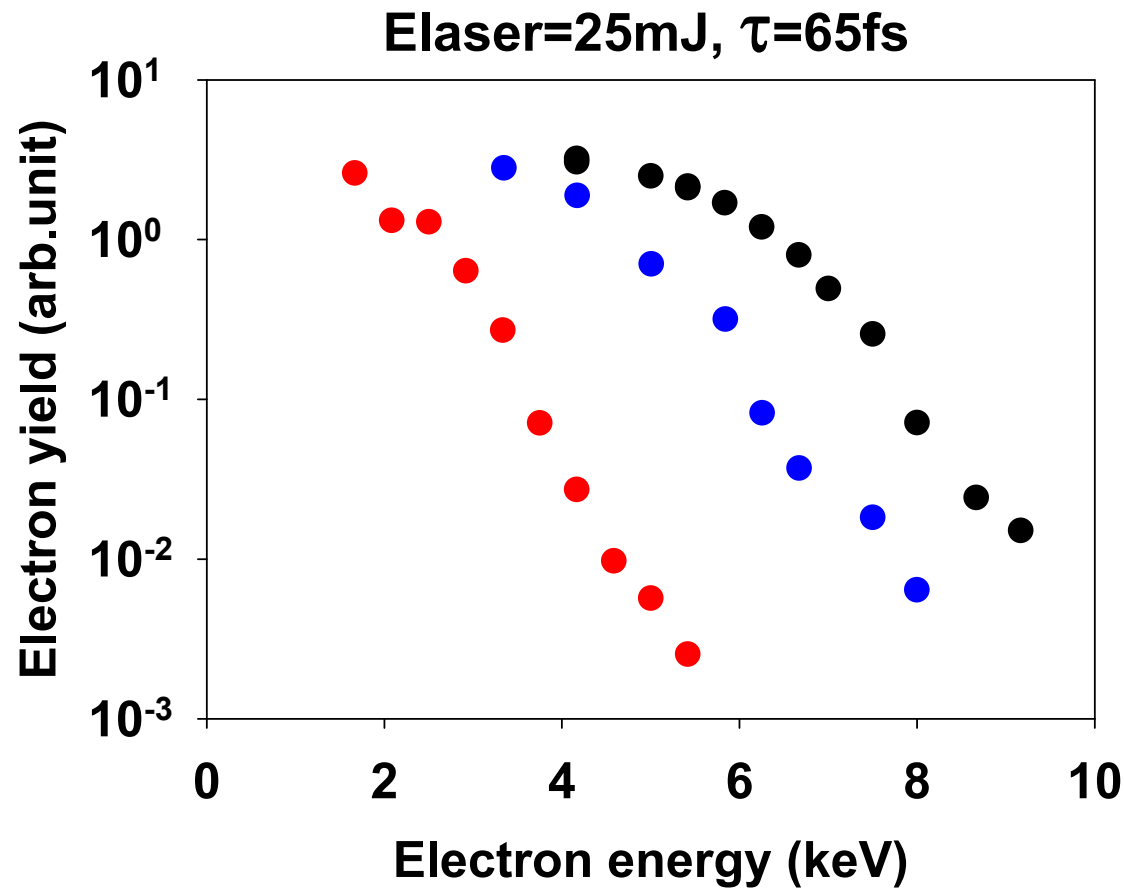


at a given $\tau=85$ fs

$N=4 \cdot 10^4$ atoms
 $I=5.3 \cdot 10^{15}$ Wcm⁻²

$U_p=12$ a.u.



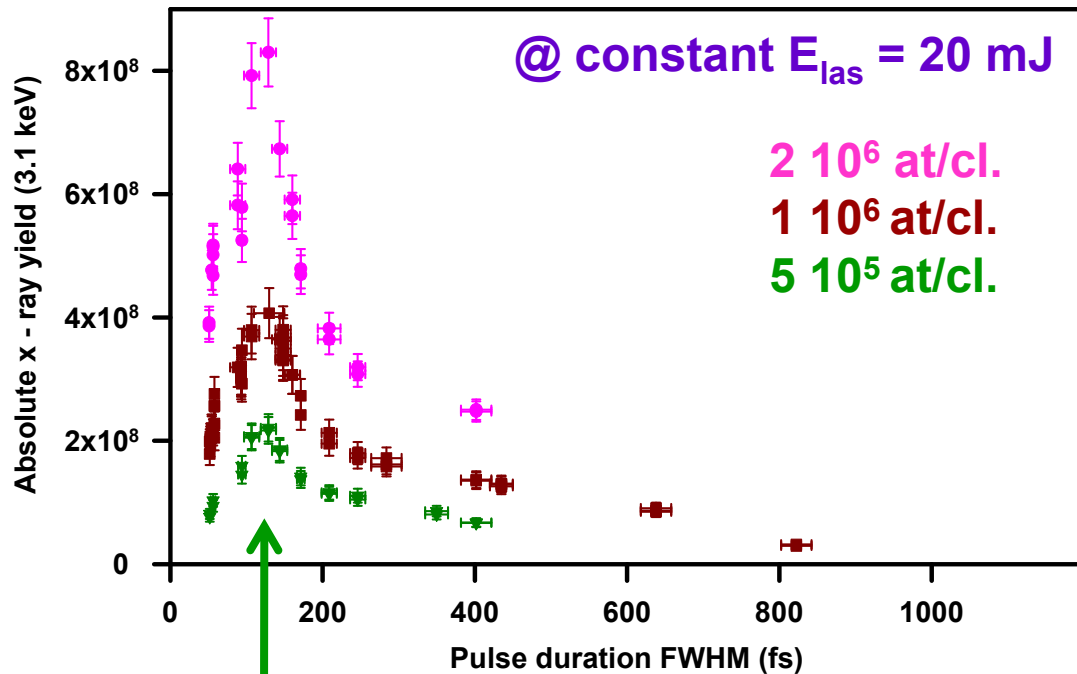


Vertical polarization: electrons are collected in the direction of the laser polarization
Horizontal polarization: electrons are collected in the direction perpendicular to the polarization of the laser

Absolute X-ray yields vs τ

$(Ar)_n : 4 \times 10^4 \text{ at/cl}, \lambda = 800 \text{ nm}, d_m \sim 10^{17} \text{ cm}^{-3}$

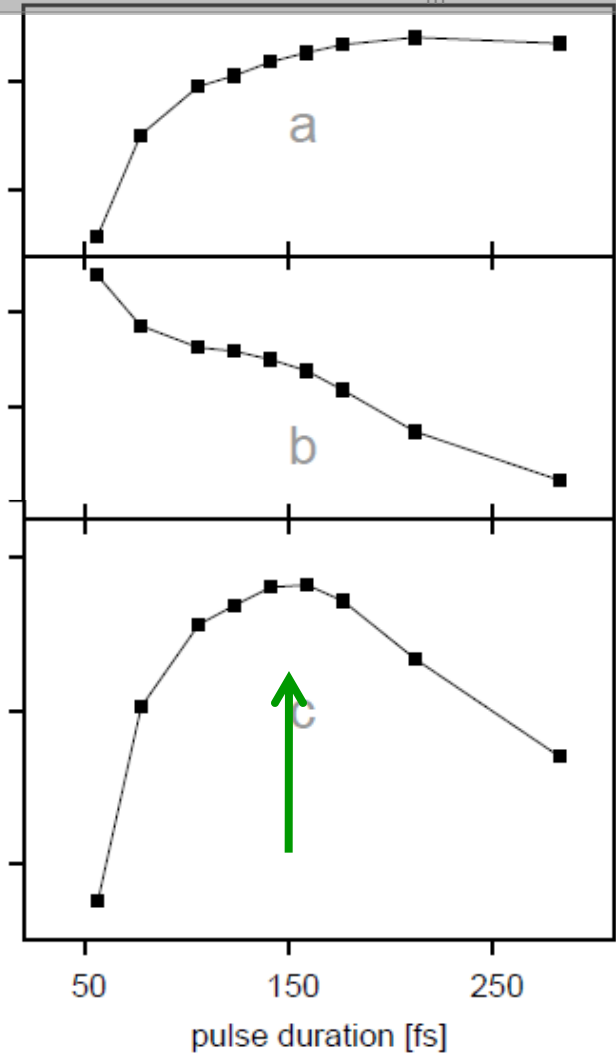
$4 \times 10^{16} \text{ W/cm}^2 \longrightarrow 3 \times 10^{15} \text{ W/cm}^2$



$10^3 P_K$

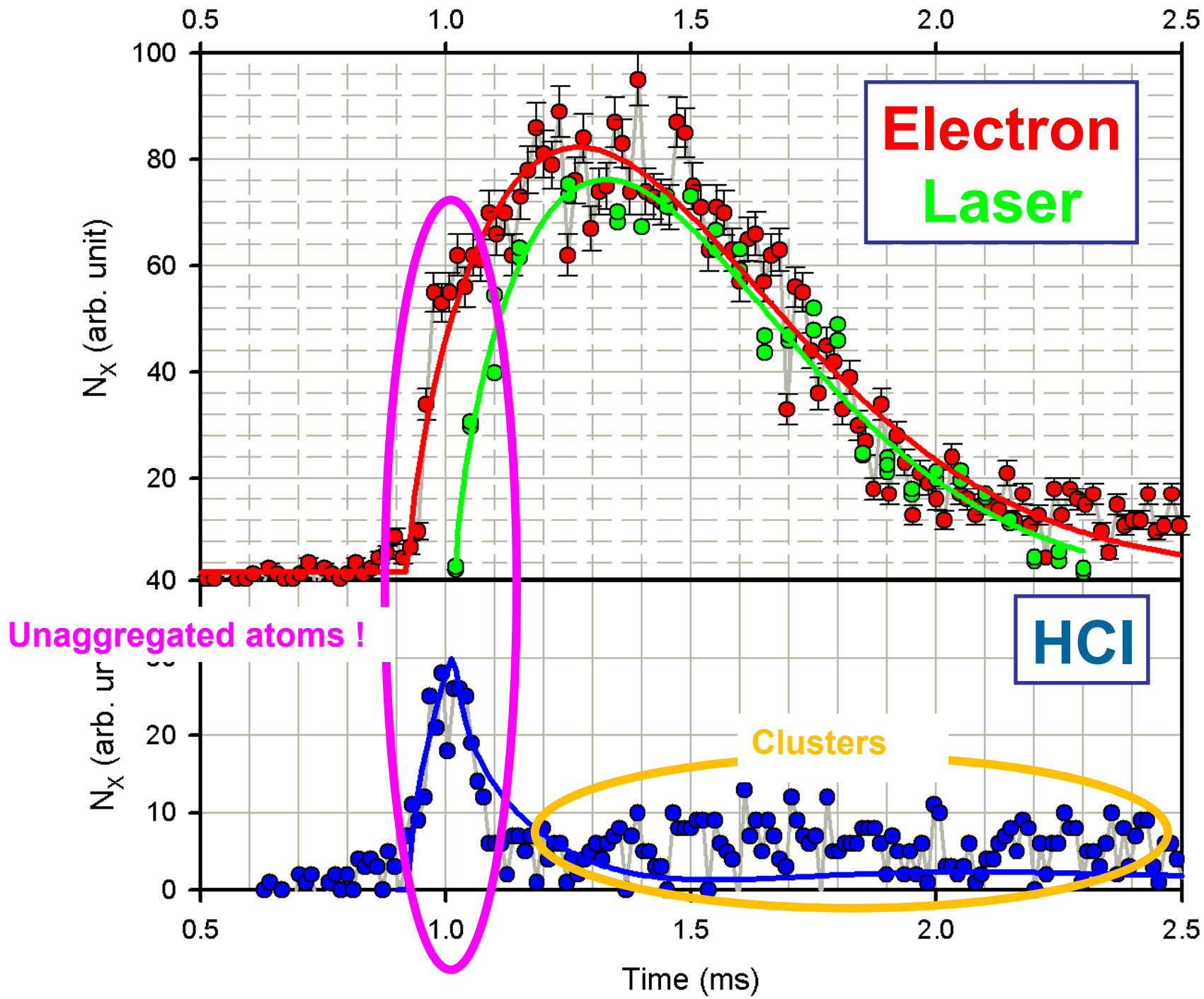
V_{eff} [arb. units]

$10^3 V_{\text{eff}} P_K$ [arb. units]



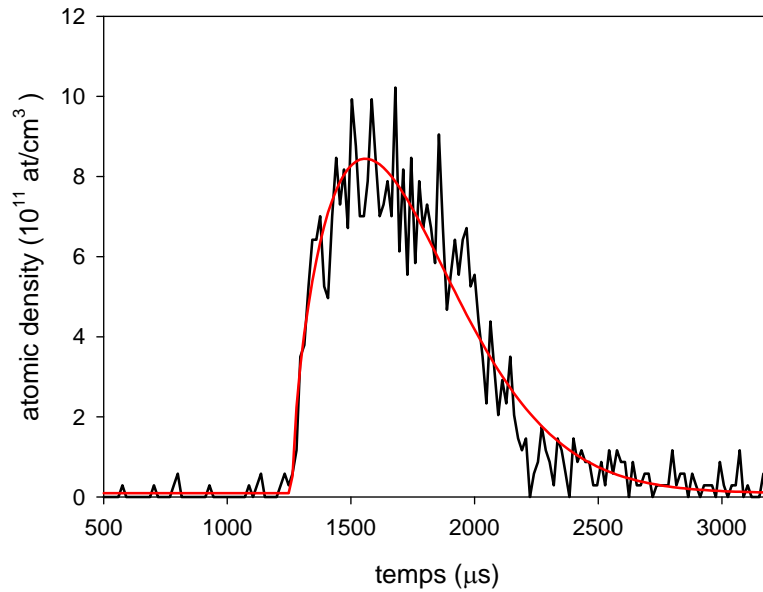
$$N_X \propto P_K(I_{\text{peak}}, \tau) \times V_{\text{eff. foc.}}(I_{\text{peak}}, \tau)$$

competition between $N_{X/cl.}$ (via P_K) and $n_{cl.}$ (via $V_{\text{eff. foc.}}$)



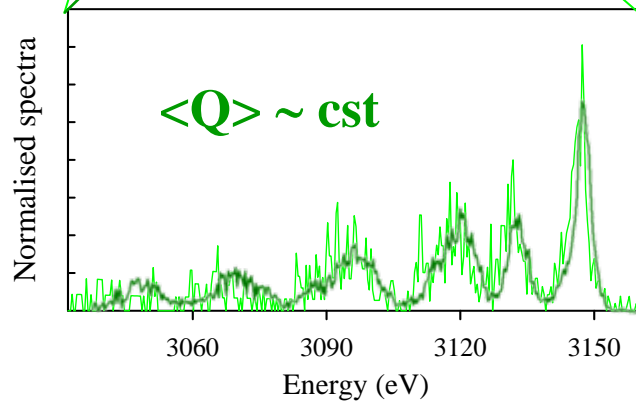
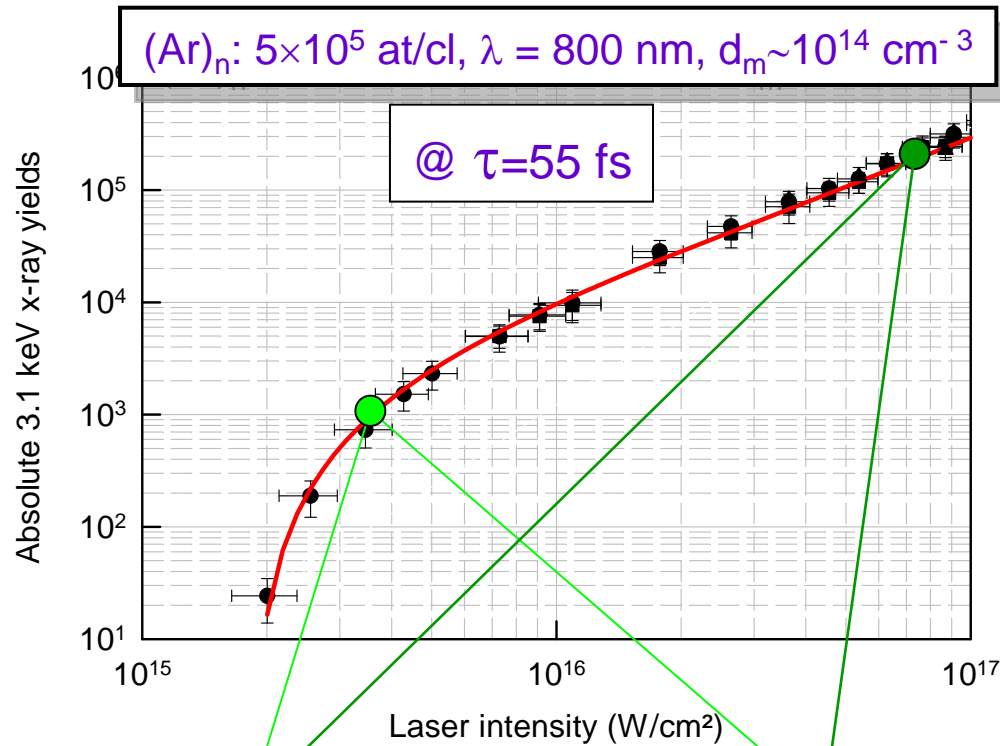
Better knowledge on the cluster jet as the atomic density, the clustering rate...

(under progress)

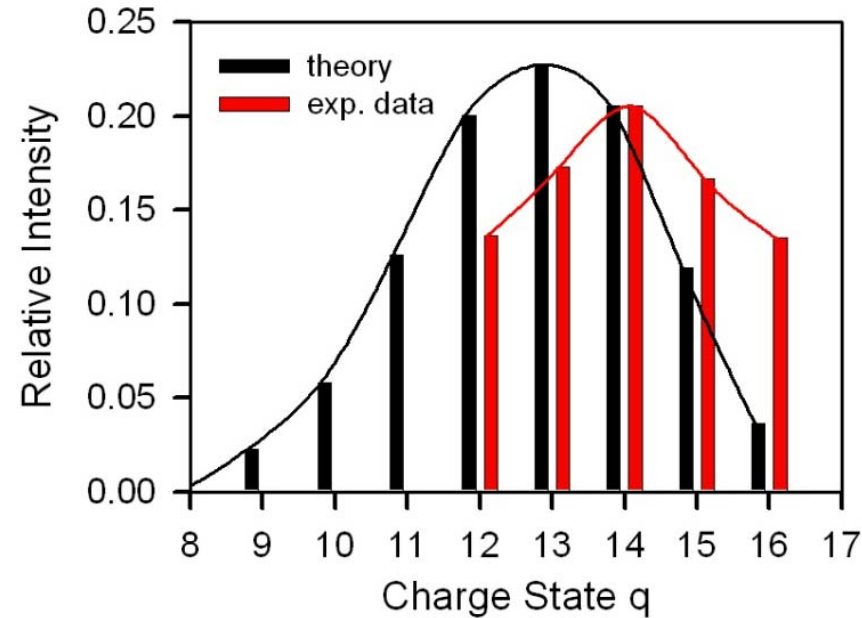


Time dependence of the cluster jet profile for $P_0 = 20$ bars, an opening time of the valve of 500 μ s and at a distance of 396 mm.

Charge state distributions



C. Prigent, C. Deiss *et al* PRA **78** (2008)



rather good agreement
for $I > 10^{16}$ W/cm²
exp. & th. differ only by one q

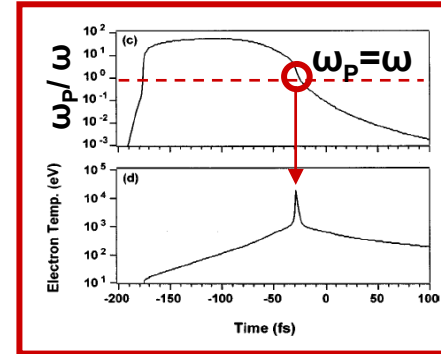
short laser - cluster interaction

The main theoretical descriptions:

► Nanoplasma model

T. Ditmire et al., Phys. Rev. A 57, 369 (1999)

revisited by F.Megi et al., J. Phys. B, 36, 273 (2003)



- tunnel ionization (W_{tun}) + collisional ionization (W_{coll})
- electron oscillation due to E_{int} + Inverse Bremsstrahlung (*via* $v_{\text{électronique}}$)
- Cluster explosion *via* coulomb effect and hydrodynamic pressure

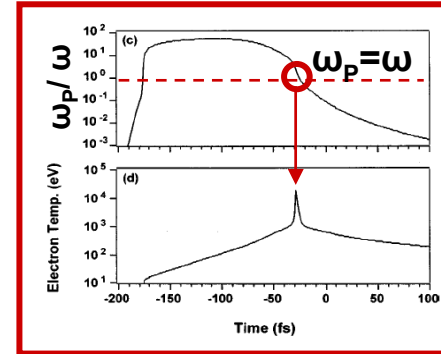
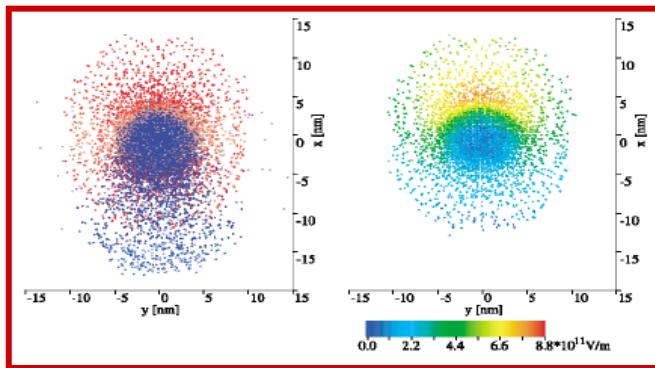
resonance: $n_e = 3 \cdot n_c \Rightarrow$ amplification of E_{int}

short laser - cluster interaction

The main theoretical descriptions:

► Nanoplasma model

T. Ditmire et al., Phys. Rev. A 57, 369 (1999)
revisited by F. Megi et al., J. Phys. B, 36, 273 (2003)



► Molecular dynamics simulations - MPIC

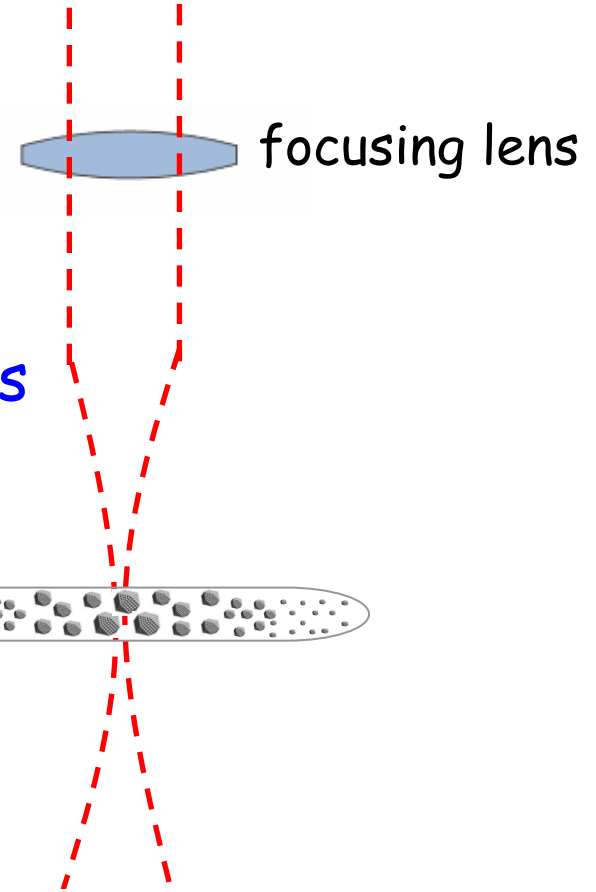
K. Ishikawa and Th. Blenski , PRA 62, 063204 (2000)
C. Jungreuthmayer et al., PRL 92, 133401 (2004)

- field ionization + electronic impact ionization
- Calculation of the electric field around each particle;
electron and ion motions are followed

the role of cluster polarization
for the production of fast multicharged ions

Experimental method

Control of the spatial and temporal overlap
between
the laser pulse and the cluster bunch



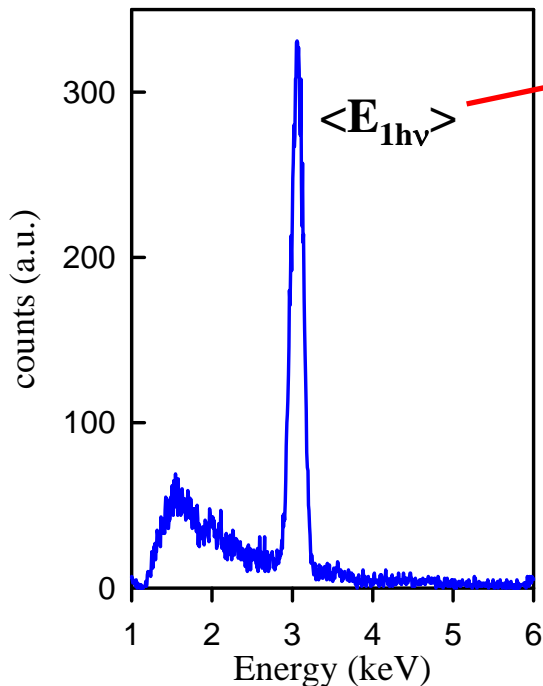
reproducible experimental conditions
shot by shot

(one laser shot for one cluster bunch)

Optimisation of the X-ray signal

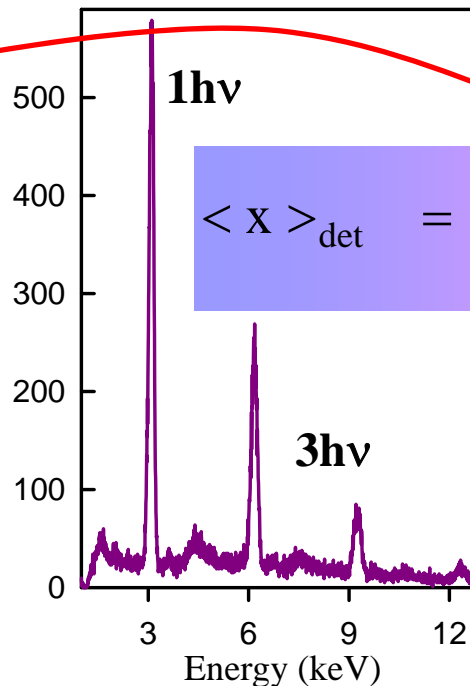
Two semiconductor detectors : make use of pile-up process

Counting rate $\ll 1$ hv/shot



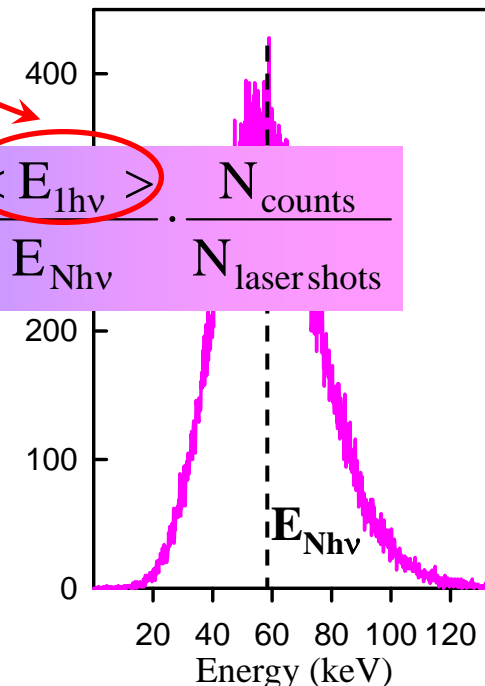
Single photon spectrum

~ 1 hv/shot



Poisson's distribution

$\gg 1$ hv/shot

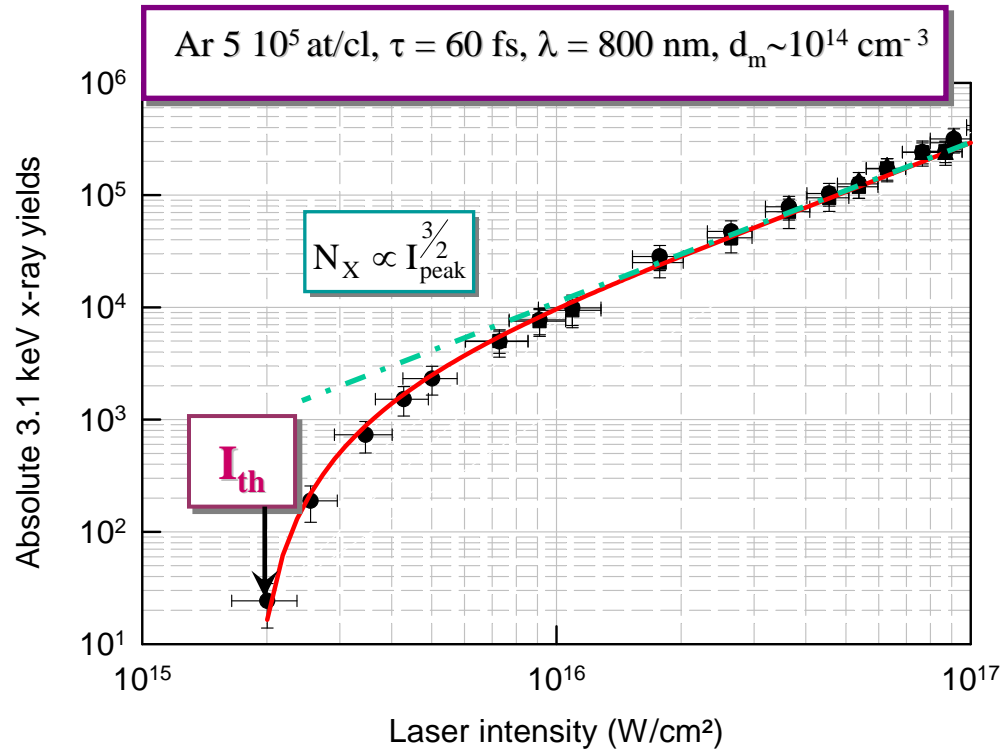


Gaussian distribution

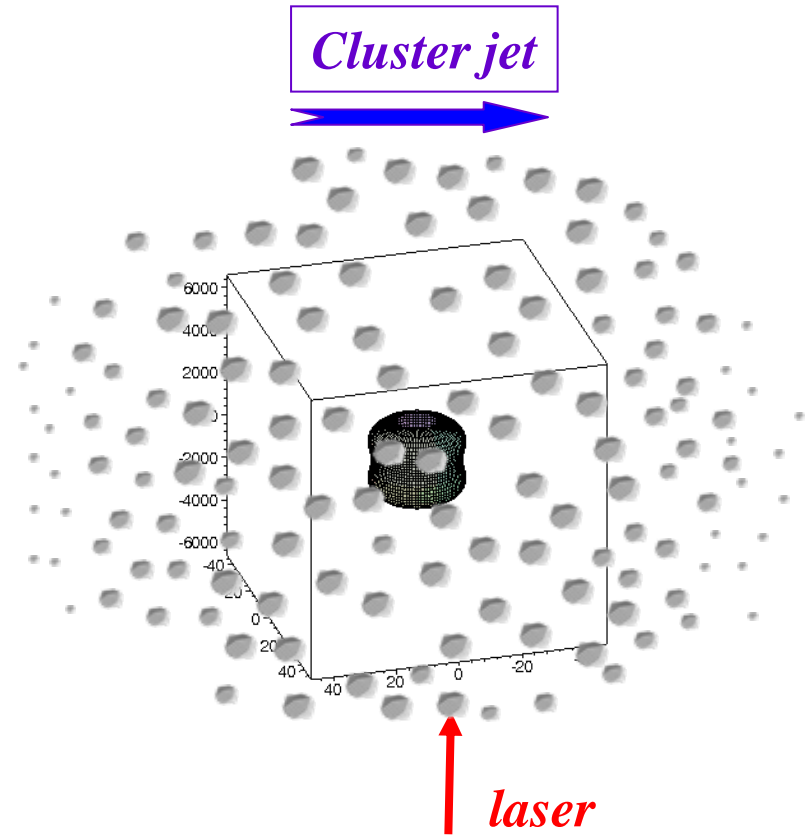
$$\langle X \rangle_{\text{det}} = \frac{\langle E_{1hv} \rangle}{E_{N_{hv}}} \cdot \frac{N_{\text{counts}}}{N_{\text{laser shots}}}$$

Absolute x-ray yields over 4 orders of magnitude
 example: from 10^5 up to $2 \cdot 10^9$ photons/laser shot in 4π (argon clusters)

Evolution with the laser intensity



Lamour, Prigent, Rozet, Vernhet NIMB **235** (2005)
 Deiss, Burgdörfer et al PRL **96** (2006)



$$V_{\text{eff foc}}(w_0, \lambda, I_{\text{laser}}, I_{\text{th}}) \neq V_{\text{nom foc}}(w_0, \lambda)$$

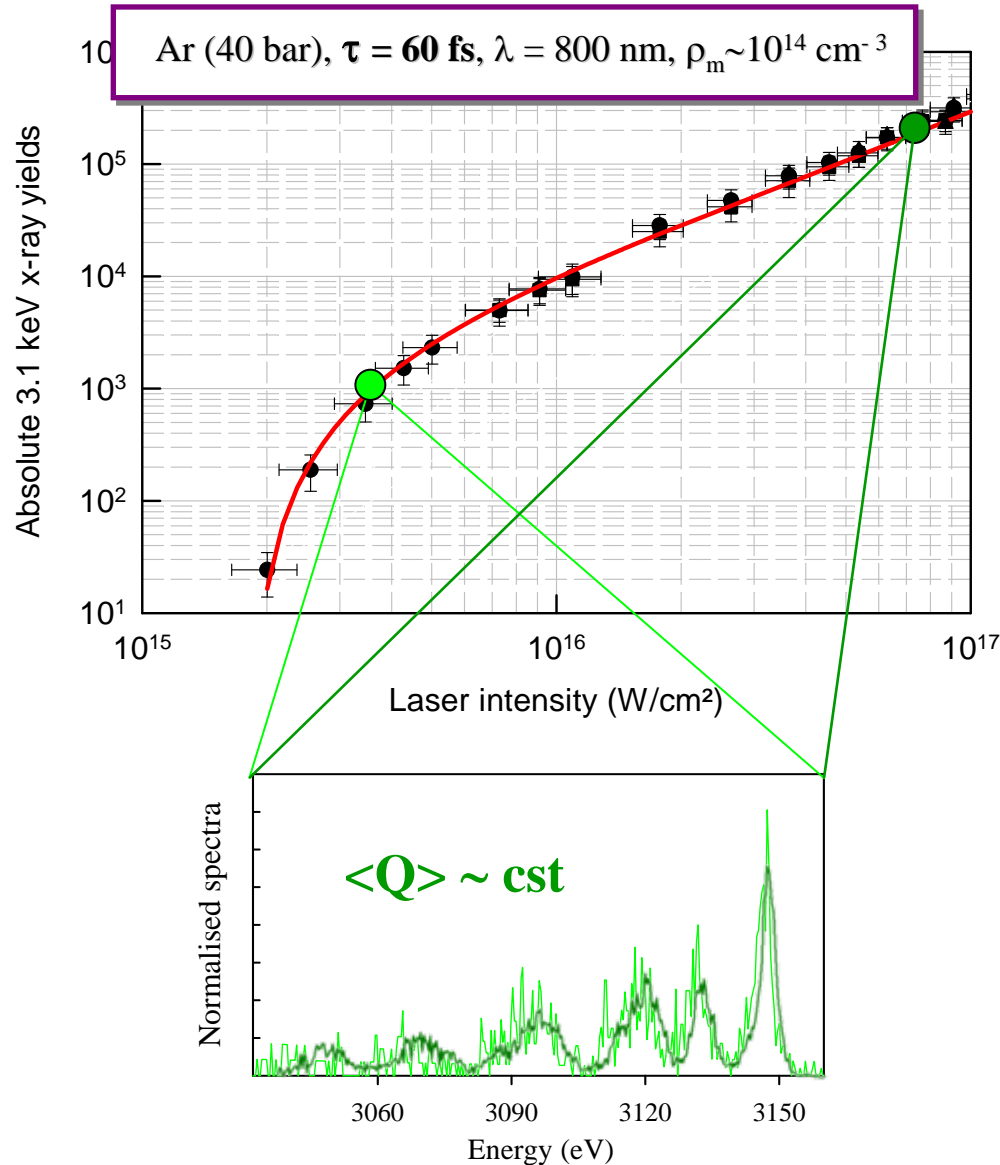
a direct outcome for $I_{\text{laser}} > I_{\text{th}}$ Pionisation inner shell $\approx \text{cst}$

$$V_{\text{eff.foc.}} = \frac{4}{3} \cdot \frac{\lambda^3}{I_{\text{th}}} \cdot \left(\frac{I_{\text{peak}}}{I_{\text{th}}} - 1 \right)^{3/2}$$

from $3 \cdot 10^{-4}$ to 10^{-3} for (Xe) $_n$

$\sim 10^{-5}$ for (Ar) $_n$

Evolution with the laser intensity



$$N_x \propto V_{\text{eff foc}}$$

✓ a direct outcome

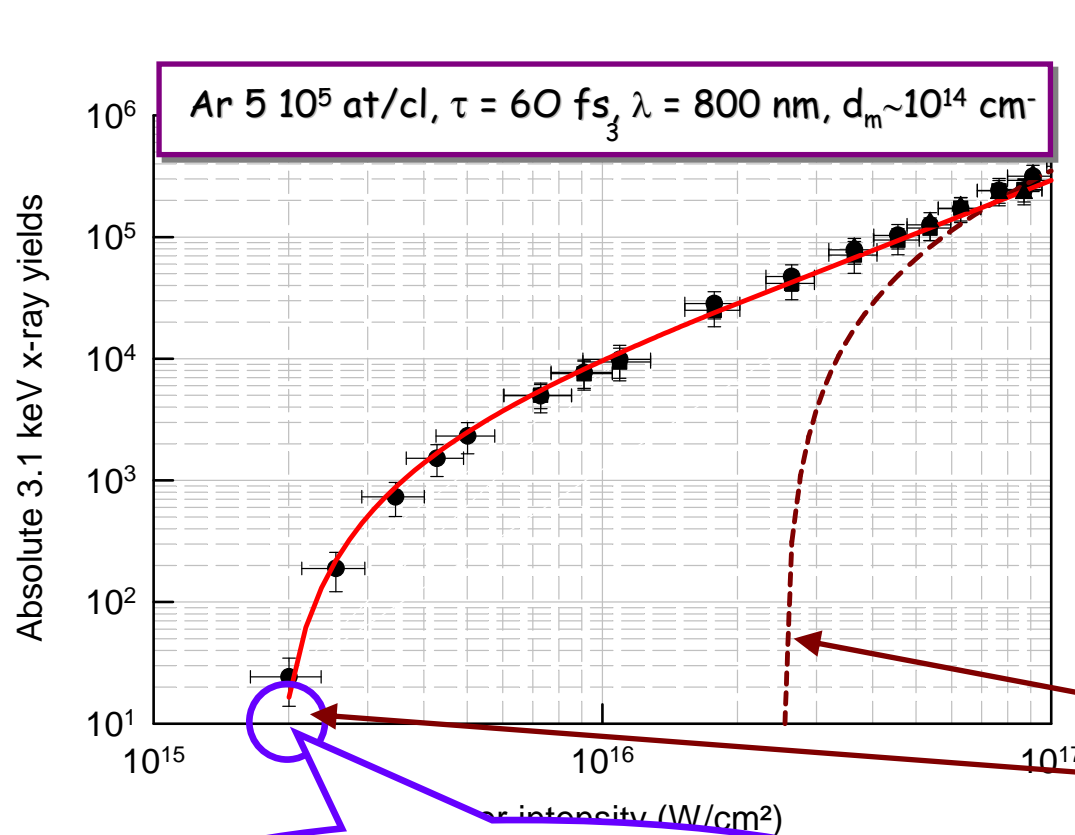
$$P_{\text{x-ray/at}} = \frac{N_x}{\langle n \rangle_{\text{eff}}} \approx \text{constant}$$

with $\langle n_{\text{eff}} \rangle = \rho \times V_{\text{eff.foc}}$

✓ In addition :

Saturation of the ionisation probabilities
for L and K shells

Absolute X-ray yield versus laser intensity



Exp.: $\sim 2 \cdot 10^{15}$ W/cm²

$I_{th}??$
the "heating" processes?

"Optical Field Ionisation"
Ar¹⁶⁺ (1s) $\rightarrow 4.0 \cdot 10^{21}$ W/cm²

Nanoplasma model

Ditmire *et al.*, Phys. Rev. A **53** (1996)

Megi *et al.*, J. Phys. B **36** (2003)

$I < 10^{16}$ W/cm² $\rightarrow T_e < 80$ eV

Ponderomotive energy

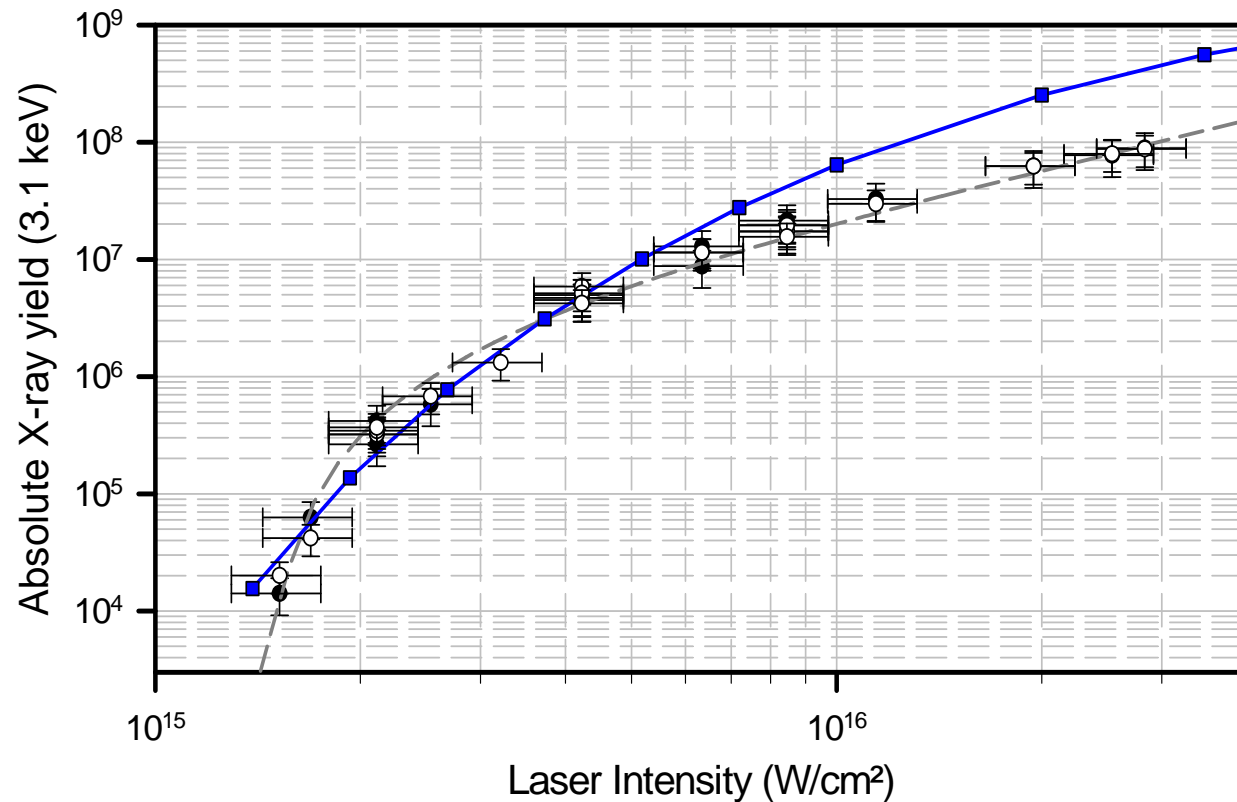
$2U_p = 3$ to 4 keV $\rightarrow 2$ - $3 \cdot 10^{16}$ W/cm²

$2U_p = 260$ eV $\leftarrow 2 \cdot 10^{15}$ W/cm²

additional heating mechanisms for electrons in clusters?

Comparison with experiment: N_x versus I

Ar (40bar): $\langle N \rangle = 5 \cdot 10^5$ at/cl, $\tau = 60$ fs, $\lambda = 800$ nm, $d_m \sim 10^{14}$ cm $^{-3}$



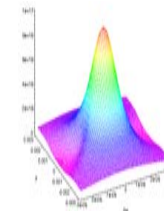
fluorescence yield

$$\omega_f = 0.25$$

clustering rate

33%

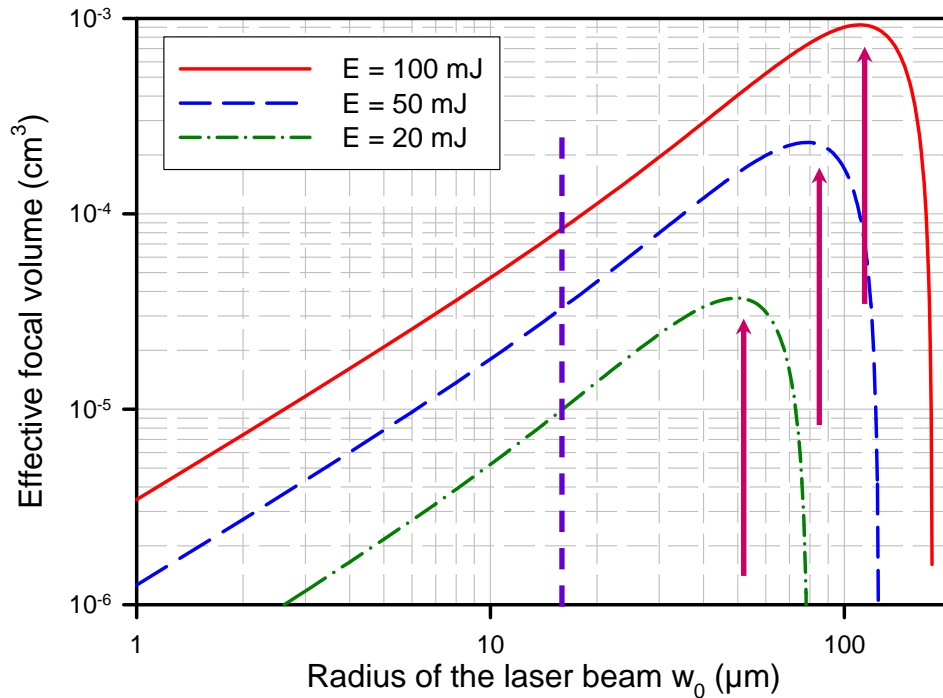
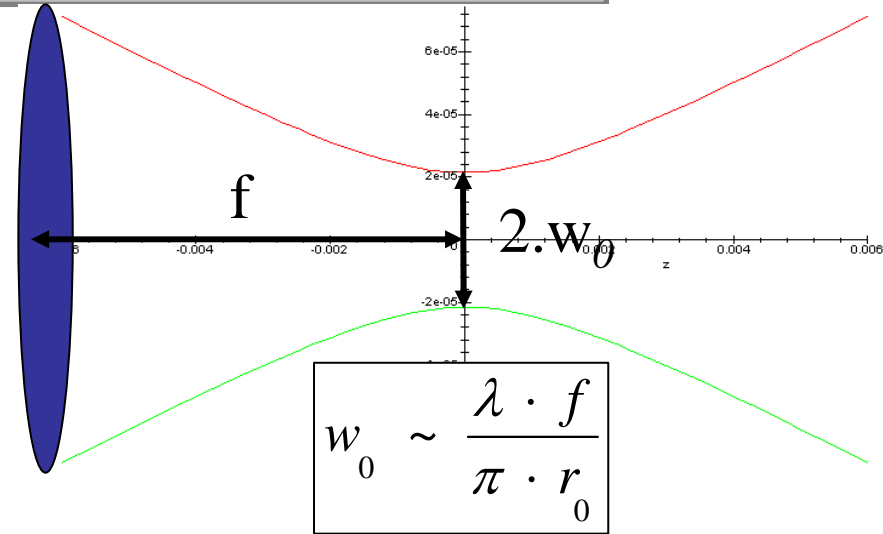
Laser intensity:
spatial distribution



Consequence on the optimization of the x-ray yield ...

$$N_X \propto V_{\text{eff foc}}(I_{\text{th}})$$

$$I_{\text{th}} = 1.4 \cdot 10^{15} \text{ W/cm}^2 \text{ for } \tau = 65 \text{ fs}$$

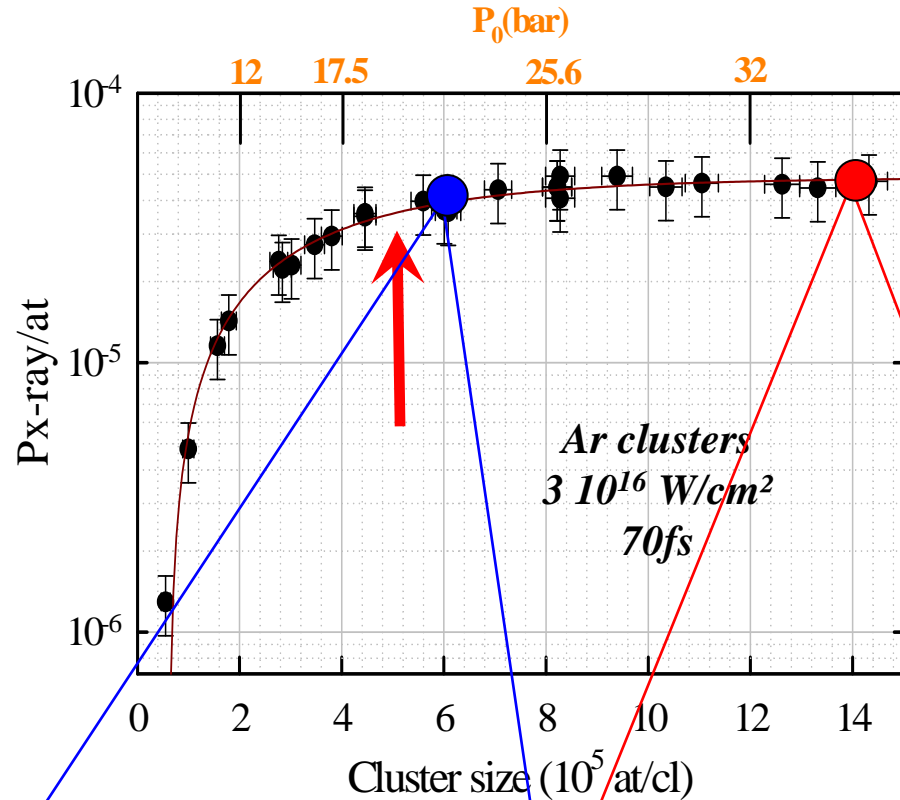


max. X-rays \Rightarrow optimum $V_{\text{Eff. Foc.}}$
defocusing up to
a quite large w_0 !!

(in our experiment : $w_0 \sim 16 \mu\text{m}$)
up to 10^9 hv/pulse in 4π for Ar_n
energy conversion $\sim 10^{-2}\%$

Cluster size dependence on the x-ray emission probability

Experimentally $\mathbf{P}_0 \Rightarrow \tilde{N} \propto \mathbf{P}_0^{1.8}$ and $\rho_{\text{mean}} \propto \mathbf{P}_0$



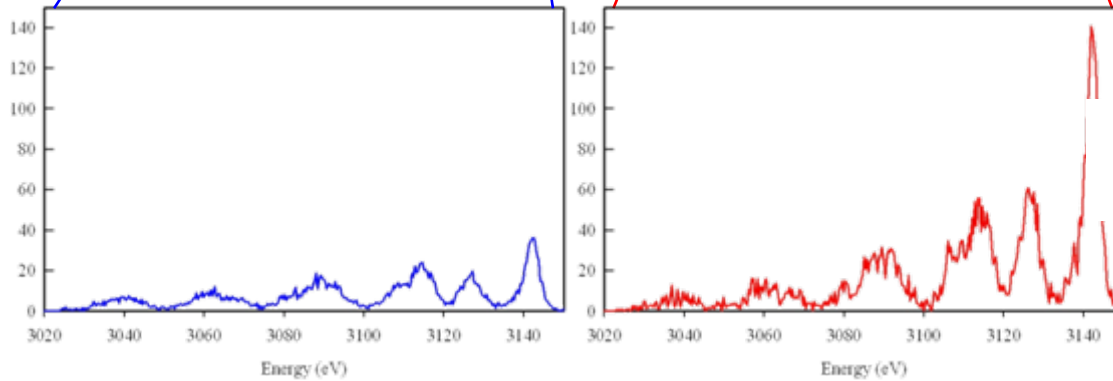
$$P_{\text{x-ray/at}} = \frac{N_x}{\langle n \rangle_{\text{eff}}}$$

with $\langle n_{\text{eff}} \rangle = \rho \times V_{\text{eff.foc}}$

$V_{\text{eff.foc}} = \text{cst}$ (whatever the size for a given τ)

Saturation of the x-ray production above a given cluster size

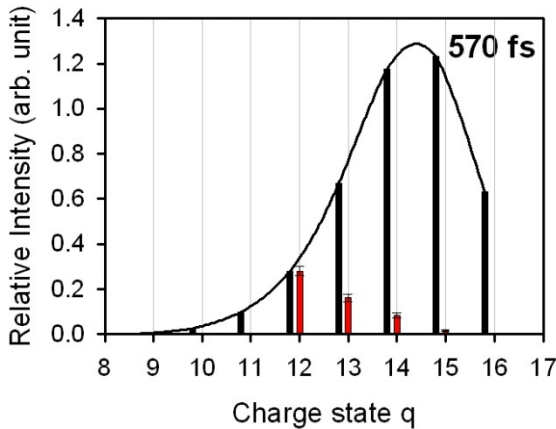
$N_x \propto P_0$ increase with the number of partners in $V_{\text{eff.foc}}$ in the saturation region



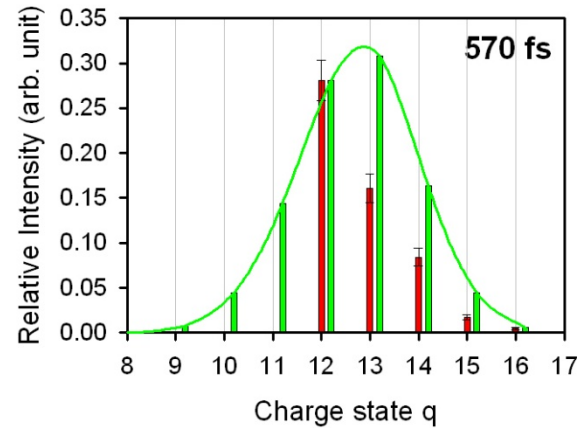
$\langle Q \rangle$ still increases somewhat \Rightarrow Increase of L shell ionisation

Comparison with the simulations: charge state distribution

Simulations with shielding



Simulations without shielding



Two limiting cases:

i) $q_i^{\text{eff}}(t)$ ionic charge reduced (shielded) by the surrounding free electrons. Particularly valid when electrons are slow (low laser intensities).

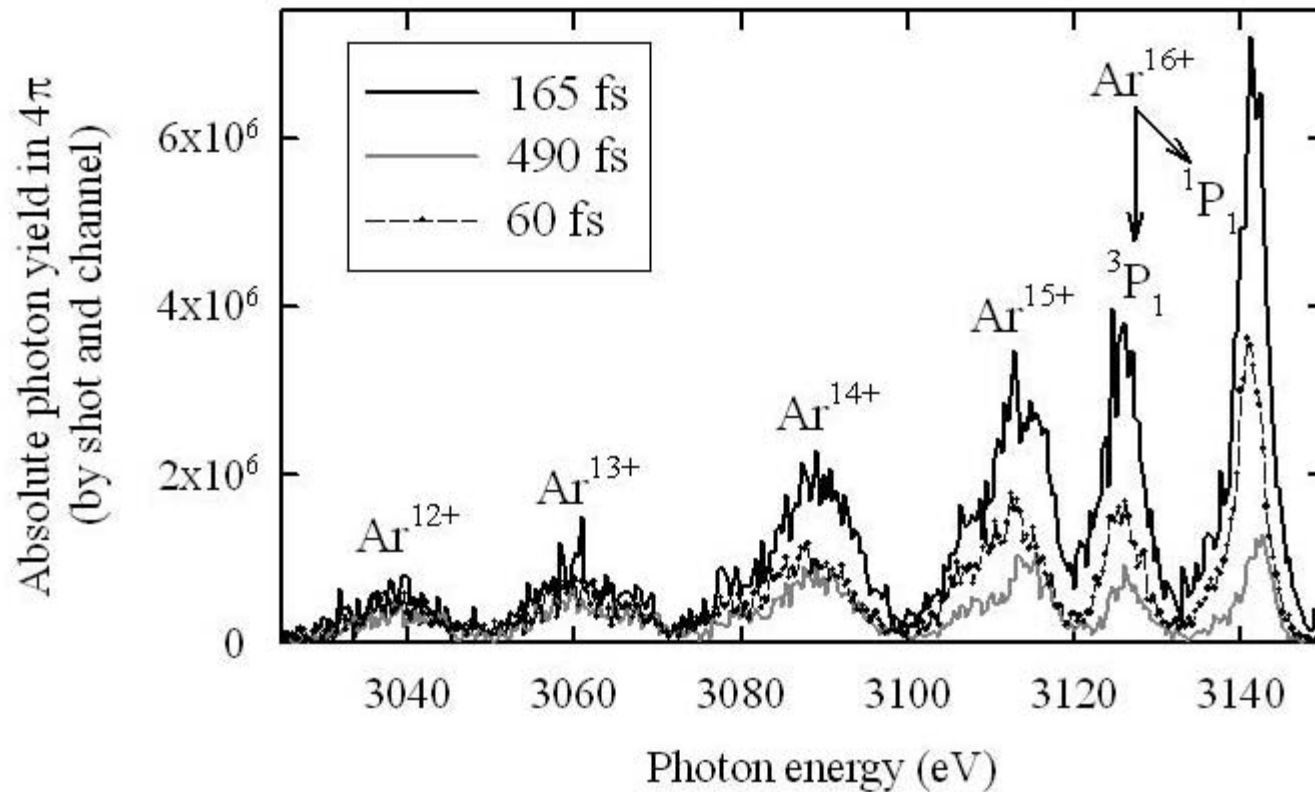
ii) $q_i(t)$ ionic charge without any shielding. Valid when electrons are too fast to be strongly influenced by ions (high laser intensities).



Influence on the description of the ion expansion

Evolution of the charge states with the pulse duration τ

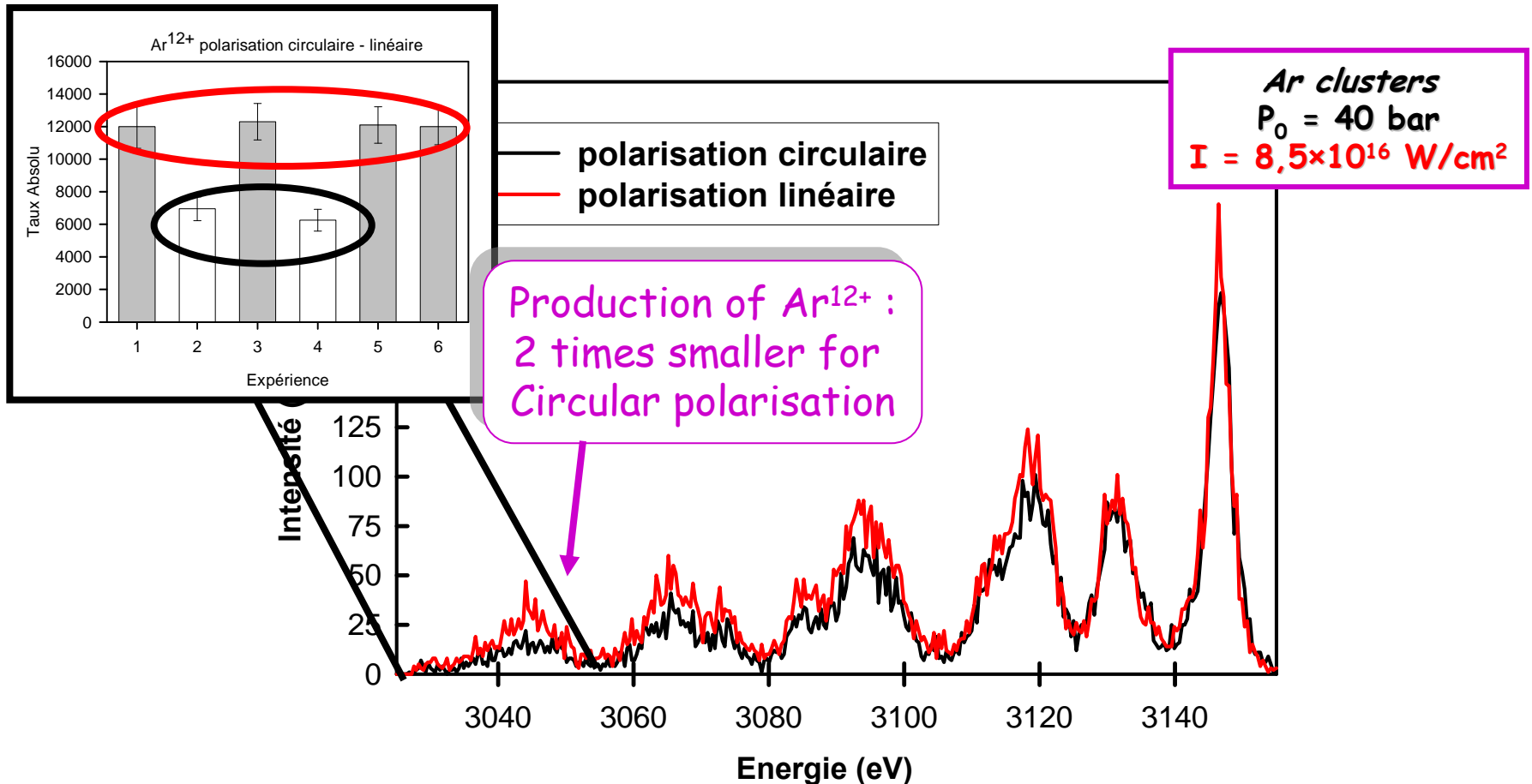
Ar $\langle N \rangle = 1 \cdot 10^6$ at/cl, $\lambda = 800$ nm, $E = 9$ mJ

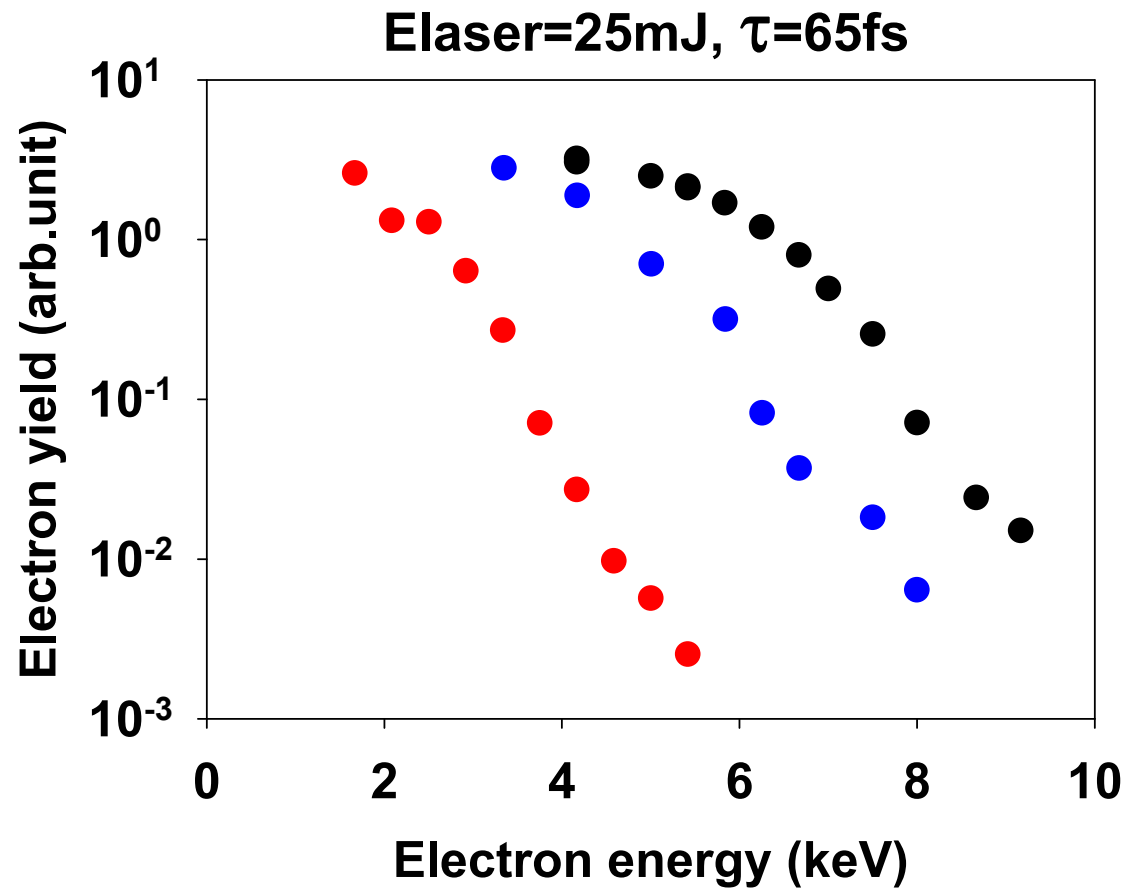


Dependence with the laser polarization

- very weak difference between linear and circular laser polarization

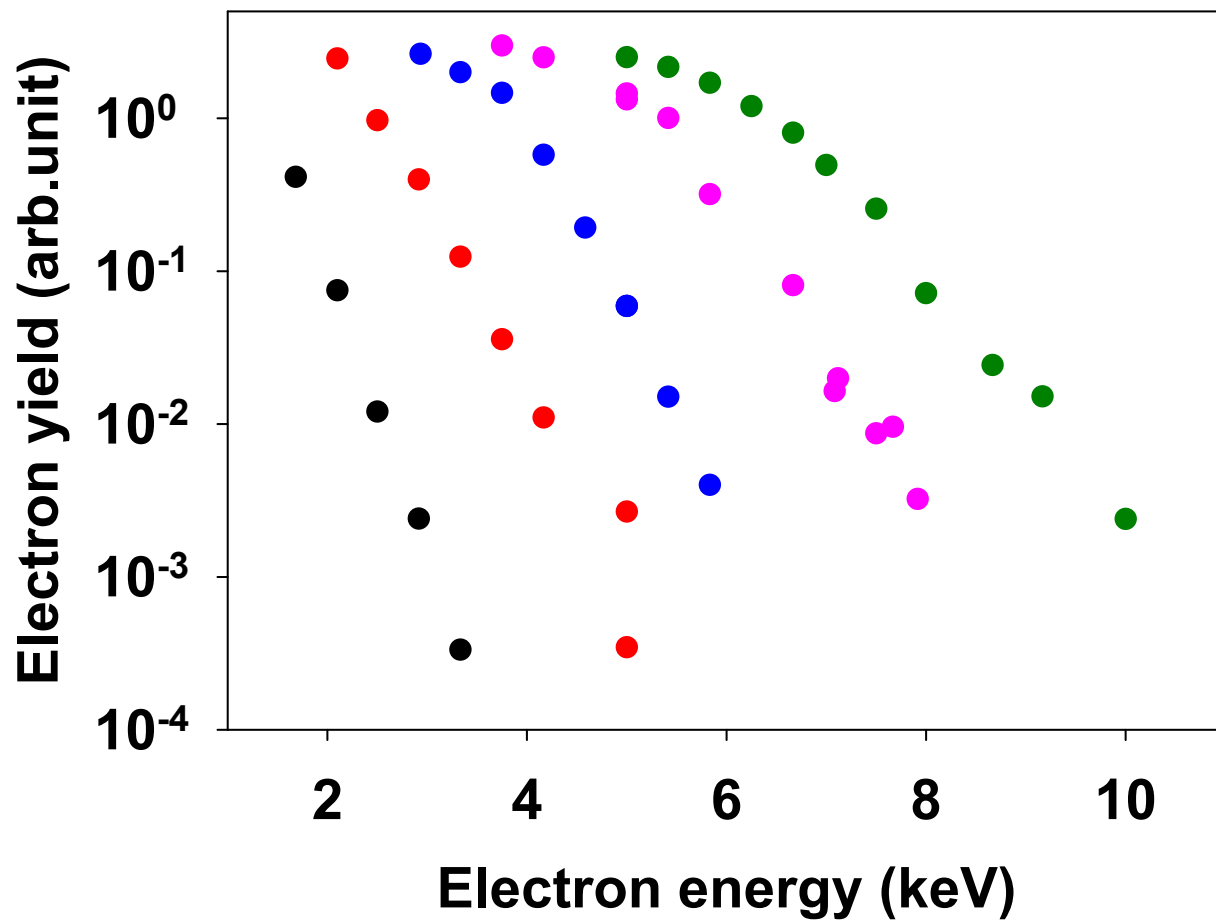
$$N_x (\text{circular}) = 80\% \times N_x (\text{linear})$$





Vertical polarization: electrons are collected in the direction of the laser polarization
Horizontal polarization: electrons are collected in the direction perpendicular to the polarization of the laser

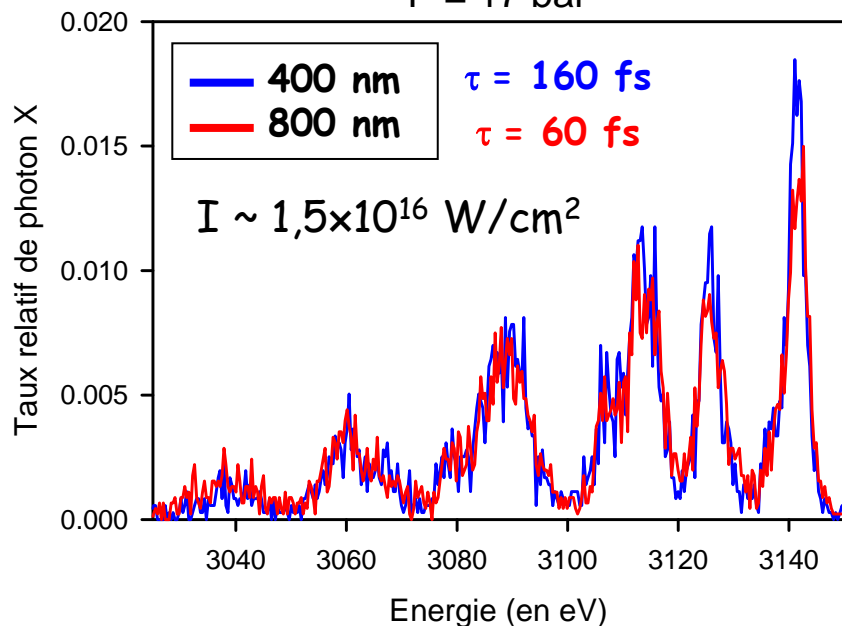
$\tau=65\text{fs}$, vertical polarization



Evolution with the laser wavelength λ (800 versus 400 nm)

Argon cluster

P = 17 bar



Same charge state distribution

For Ar or Xe

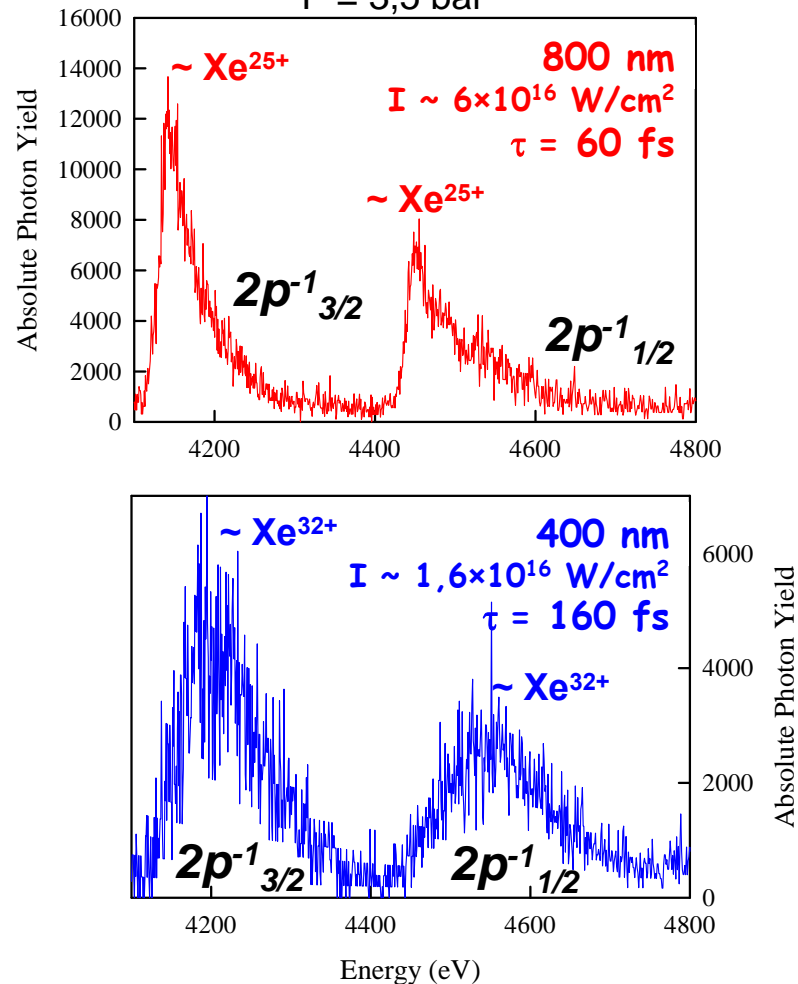
τ , I and N comparable:

$$N_X(800 \text{ nm}) \sim 10 \times N_X(400 \text{ nm})$$

~~$$N_X(800 \text{ nm}) \ll N_X(400 \text{ nm}) \text{ (QP model)}$$~~

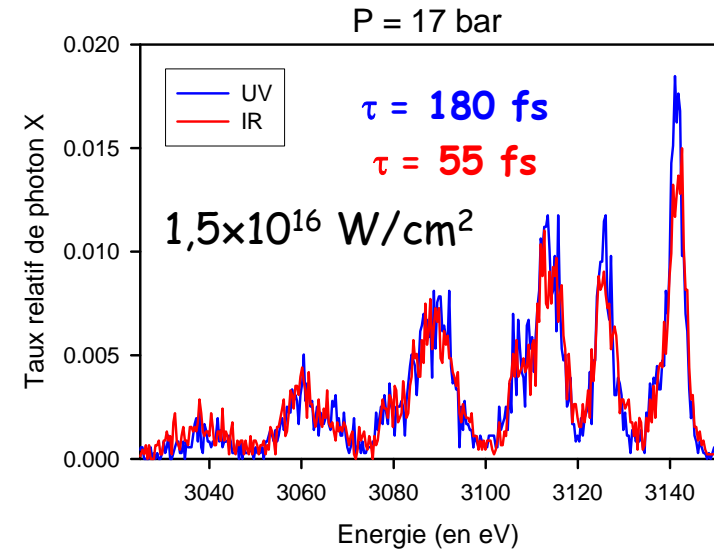
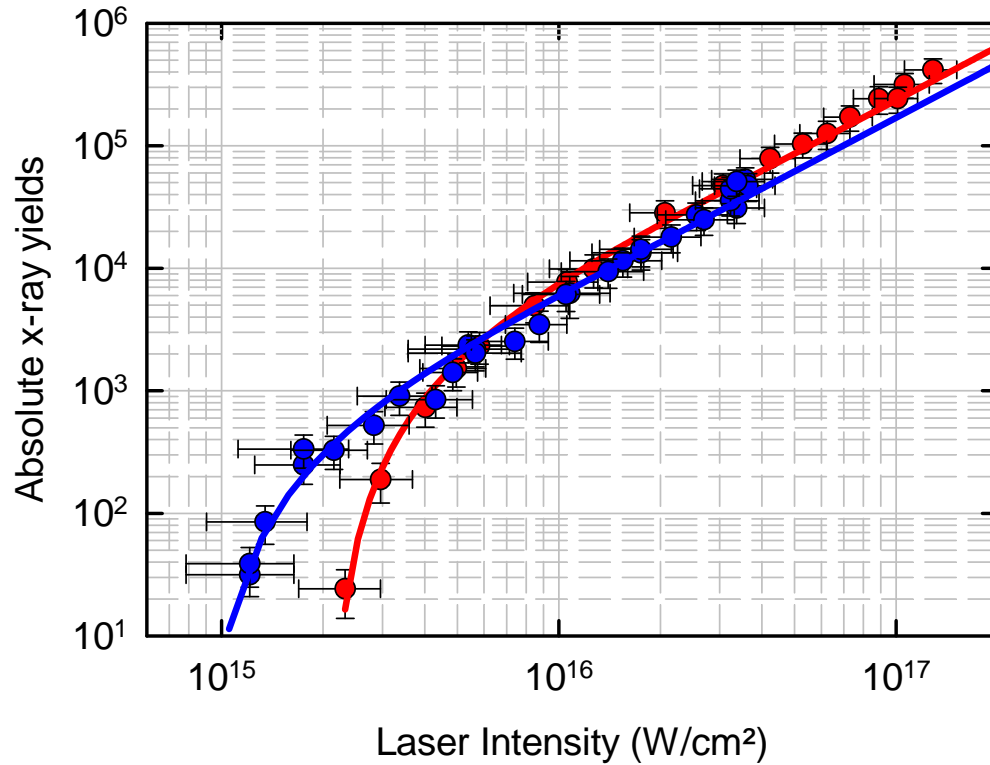
Xenon cluster

P = 3,5 bar



Different charge state distribution

Influence of the laser wavelength

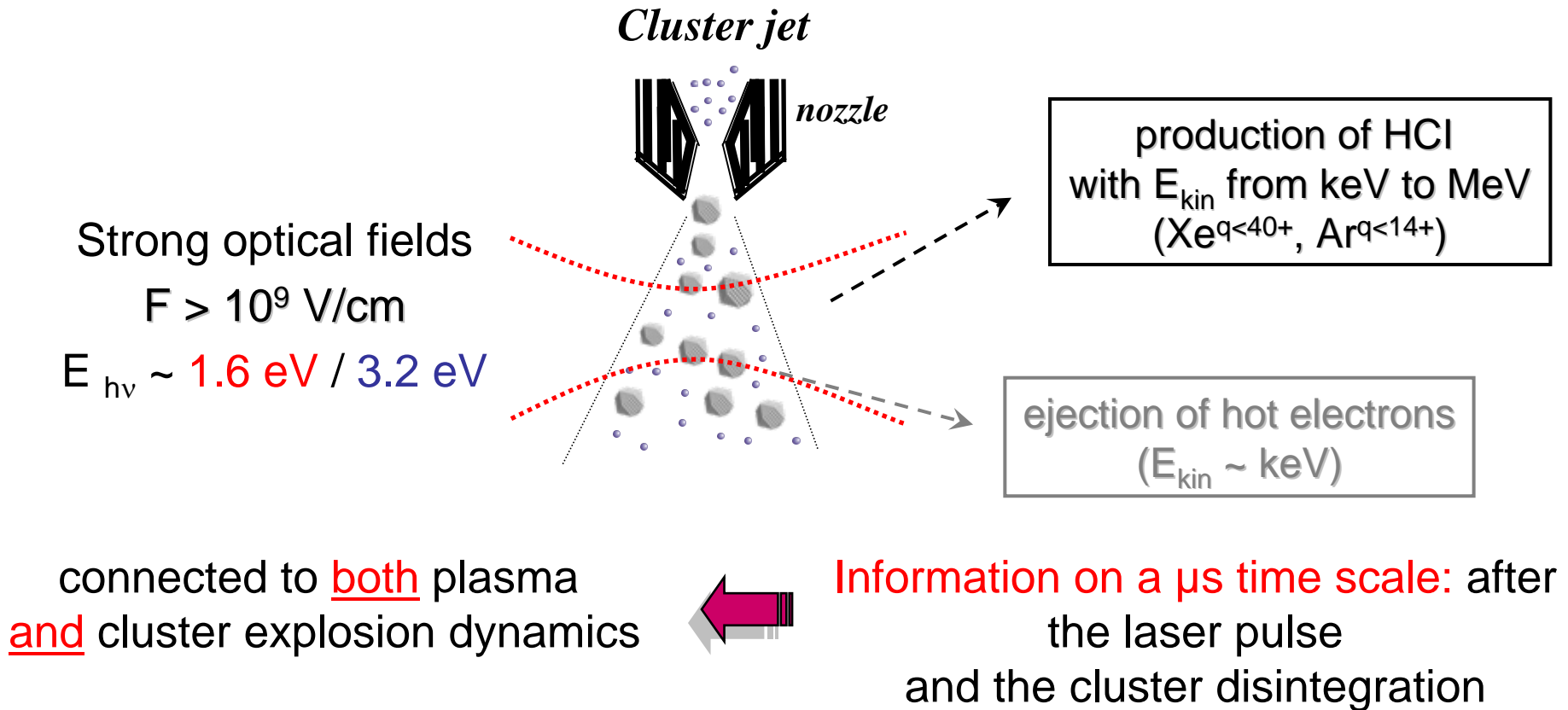


λ (nm)	τ (fs)	I_{th} (W/cm ²)
800	55	$\sim 2.2 \times 10^{15}$
400	180	$\sim 1 \times 10^{15}$

τ , I and size comparable:

$$N_x(800 \text{ nm}) \sim 10 \times N_x(400 \text{ nm})$$

Van der Waals clusters in intense laser fields



An incomplete list of several studies:

- T. Ditmire, J. Marangos, J. Tisch *et al.*, Nature **386** (1997)
- M. Vrakking *et al.*, Phys. Rev. A **68** (2003)
- D. Mathur *et al.*, Phys. Rev. A **66** (2002), A **69** (2004) ...
- H. Ueda, Y. Kishimoto *et al.*, Phys.Rev. A **67** (2003)
- E. Skopalová, J.Tisch, J.P. Marangos *et al.* PRL **104** (2010)

Scenario of the dynamics of a single cluster

charging up of the cluster & polarization of the cluster
→ a strong asymmetry of the electric field, $F_z(z)$

Field enhanced by charging of the cluster
and polarization of the cluster

Laser field screened
by polarization of the cluster

