Simulations of intense hard x-ray induced dynamics of matter

2.7.2018 – EUCALL Workshop
"Theory and Simulations of Photon-Matter Interaction"

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Theory Division

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HELMHOLTZ ASSOCIATION

CFEL SCIENCE

DESY

Outline

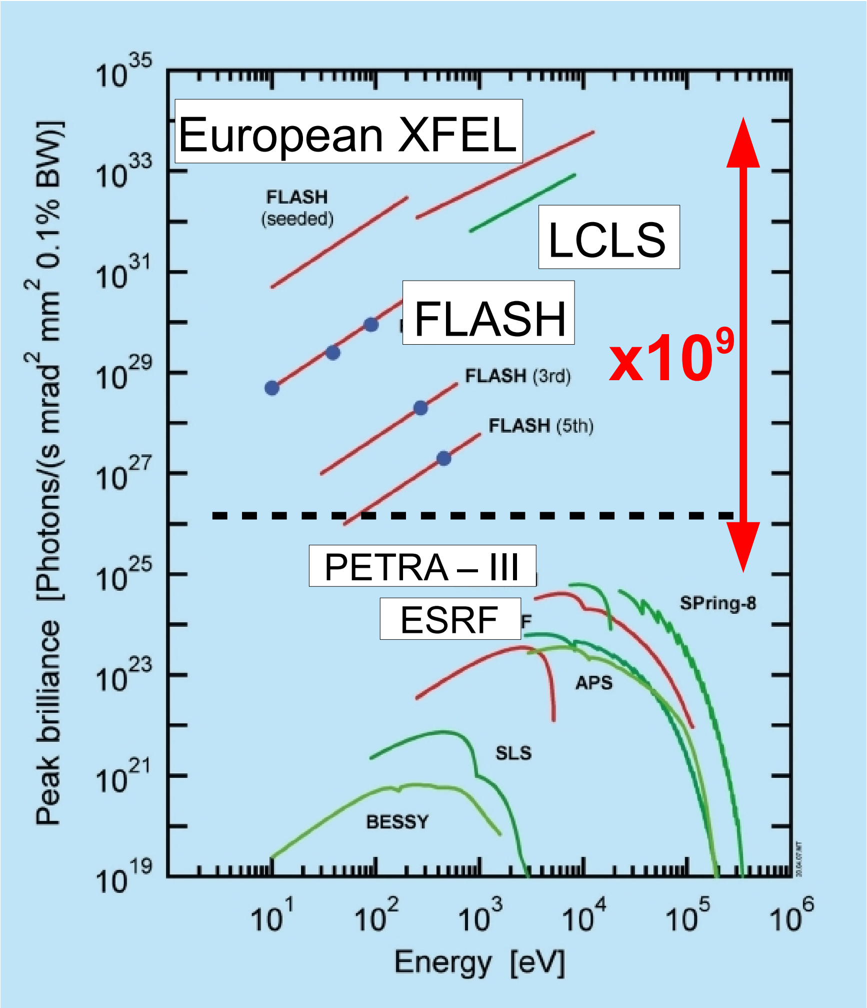
- Introduction: High intensity X-ray – matter interaction
  Challenges for modeling

- Complex dynamics of matter induced by ultra-high-intensity X rays
  – Microscopic description with the simulation tools **XMDYN** and **XATOM**
  – X-ray induced **cluster dynamics**
  – **Chemical effects** in clusters
  lower intensity, but still multiple photon absorption

Introduction

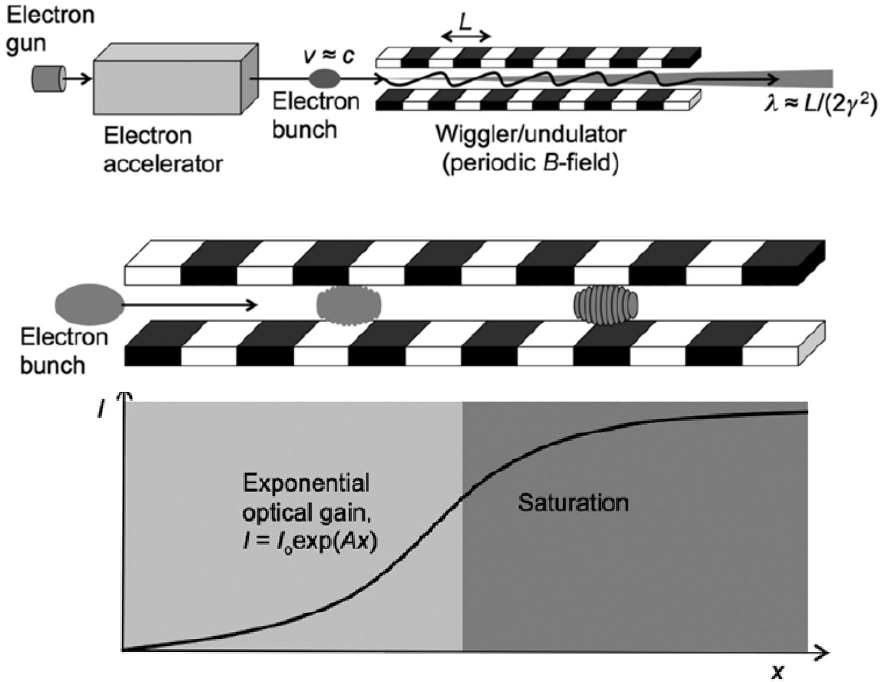
X-ray–matter interaction at high x-ray intensities

High x-ray intensity → how high is it?



photon-science.desy.de

X-ray Free-Electron Laser (XFEL)



T_{pulse} ~ 10 – 100 fs

Ribic, Margaritondo, *J. Phys. D* **45** 213001 (2012)

Pellegrini, *Rev. Mod. Phys.* **88** 015006 (2016)

High x-ray intensity → how high is it?

- **Probability of photoionization** during a single pulse (disregarding all other processes)

Fluence

probability ~ $1 - \exp(-\sigma N_{photon} / A_{focus}) = 1 - \exp(-\sigma F)$

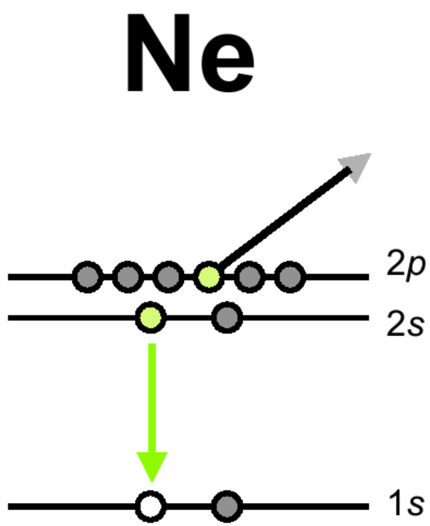
Cross section for Carbon at 1 keV: $\sigma_{Carbon} \sim 0.044$ Mb ($= 4.4 \cdot 10^{-24}$ m^2)

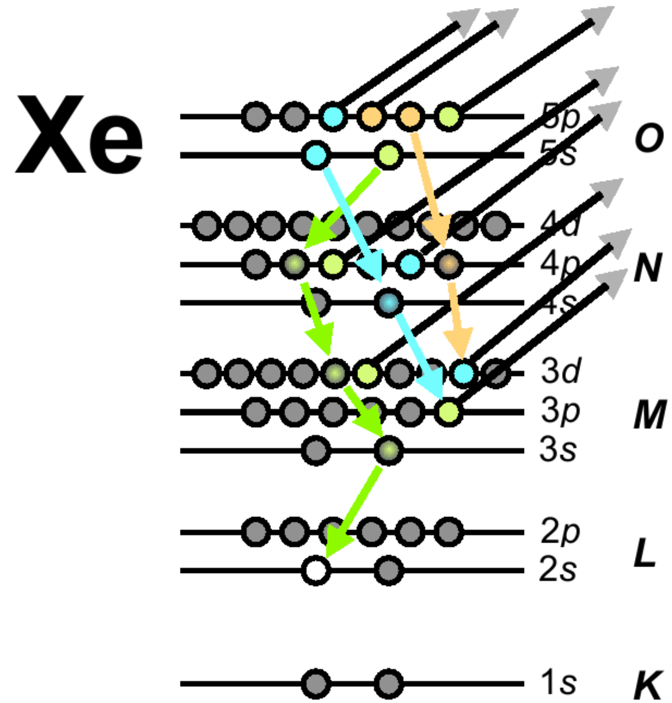
	Synchrotron	XFEL
N_{photon}/ pulse	10^6	10^{12}
A_{focus}	1μm^2	1μm^2
T_{pulse}	~ 20 ps	~ 10 … 100 fs
probability	$4.4 \cdot 10^{-5}$	**0.988**
Signal vs. Fluence	linear	**non-linear**

$1 / \sigma = F_{saturation} \leq F_{applied}$ → **High intensity**

High intensity x-ray induced dynamics: challenge for theory

> **Various** different **electronic configurations** may appear transiently





Multiphoton absorption after/during decay cascade

→ More than 20 million multiple-hole configurations

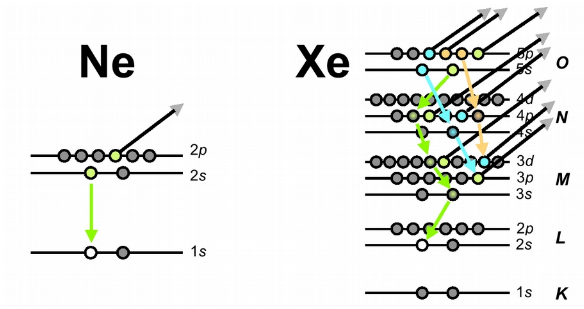
→ More than 2 billion x-ray-induced processes

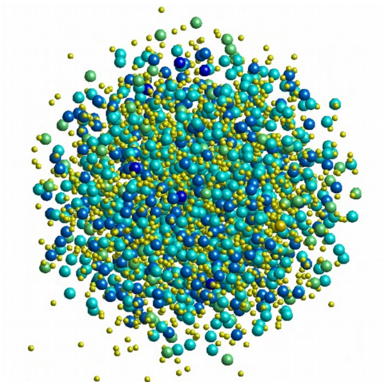
Figures courtesy of S.-K. Son

High intensity x-ray induced dynamics: challenge for theory

> **Various** different **electronic configurations** may appear transiently

In many atom systems: **environmental effects**





Non-equilibrium dynamics

> **Highly excited matter** → how to capture **theoretically**?

High intensity x-ray induced dynamics: challenge for theory

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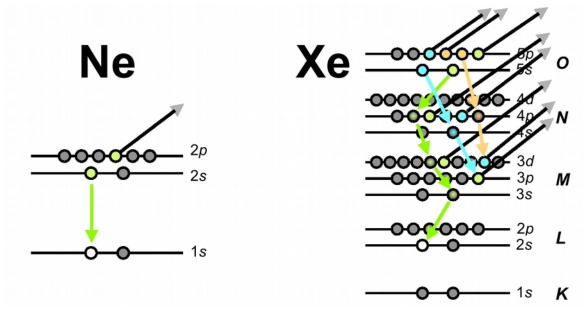
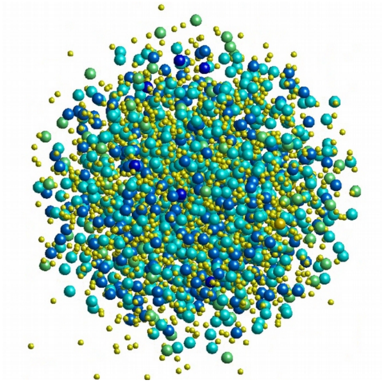
  In many atom systems: **environmental effects**





Non-equilibrium dynamics

- Our simulation tools:
  - For single atoms: **XATOM** (ab initio code)
  - For atomic clusters, many-atom systems: **XMDYN**

    (Monte Carlo / Molecular Dynamics code)

XATOM

by

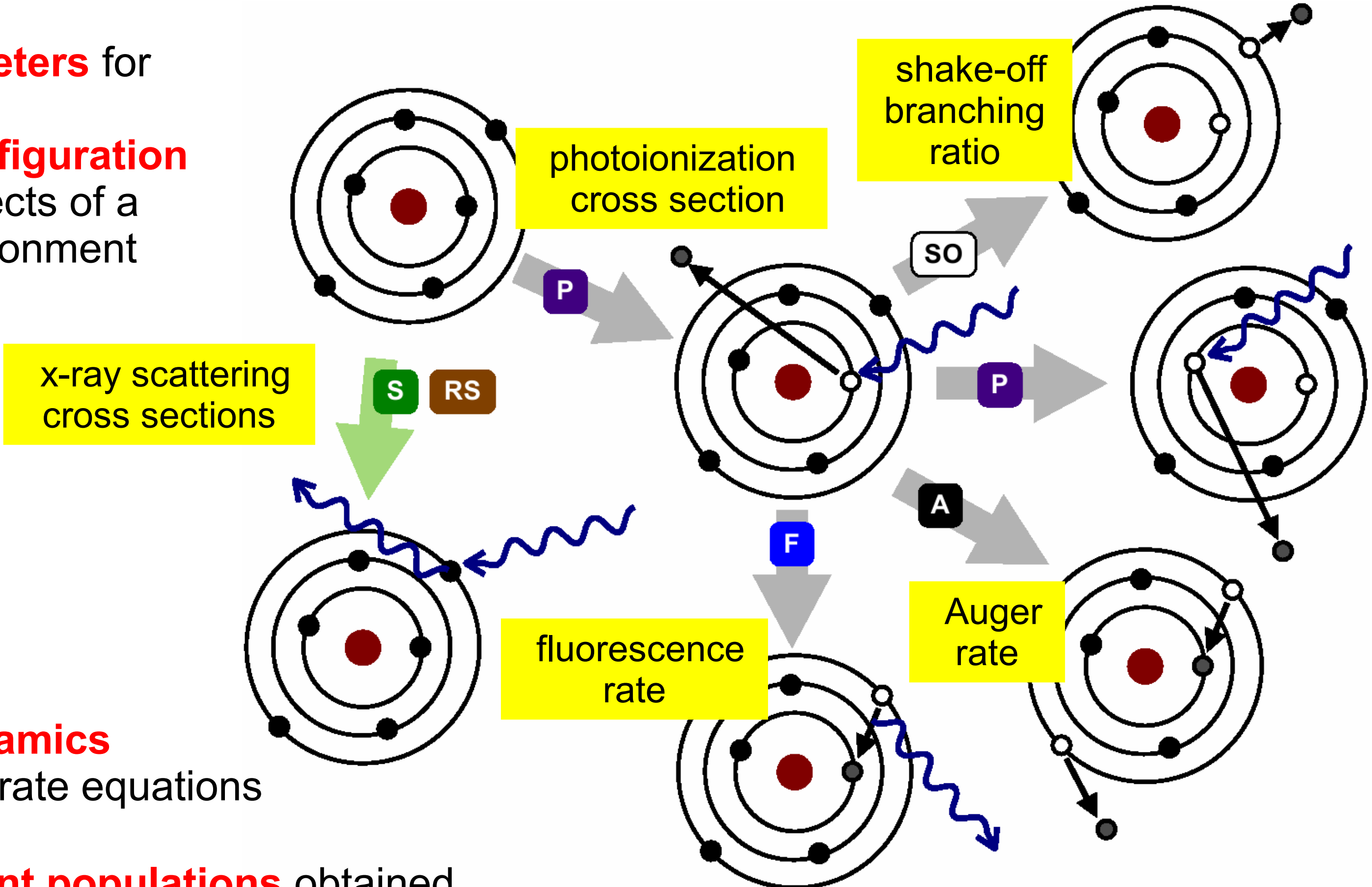
Sang-Kil Son, Jan-Malte Slowik, Koudai Toyota,

Robin Santra

(CFEL-DESY Theory Division)

XATOM: An integrated toolkit for x-ray and atomic physics

- Ab initio code based on the Hartree-Fock-Slater approach
  - **Atomic parameters** for **arbitrary electronic configuration**
    > also with effects of a plasma environment



  - **Ionization dynamics**
    > described by rate equations
  - **Time-dependent populations** obtained
    > e.g. charge state distribution

Son, Young & Santra, *Phys. Rev. A* **83**, 033402 (2011)
Jurek, Son, Ziaja & Santra, *J. Appl. Cryst*. **49**, 1048 (2016)

Dynamics at high intensities: simulation tools

XMDYN

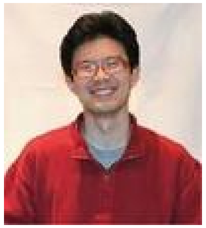
Computational tool: XMDYN

Atomistic Particle Approach + Molecular Dynamics (MD)

- Bound electrons → Occupation numbers

  Photoionization and **inner shell relaxation**: **Monte Carlo**

  Rates by **XATOM** package (Sang-Kil Son, Robin Santra)

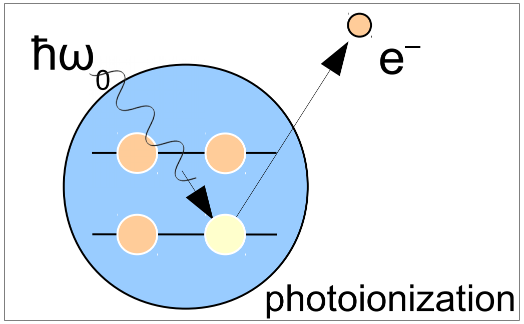


- **Real space dynamics** of atoms/ions and free electrons:

  classical **MD** → force fields (e.g. Coulomb) ; Newton's equations

- Phenomena due to the **molecular environment**

  – chemical bonds (force fields)
  – secondary (collisional) ionization
  – recombination
  – charge transfer between ionic sites



Jurek, Son, Ziaja & Santra, *J. Appl. Cryst*. **49**, 1048 (2016)
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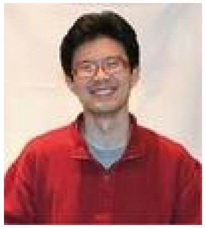

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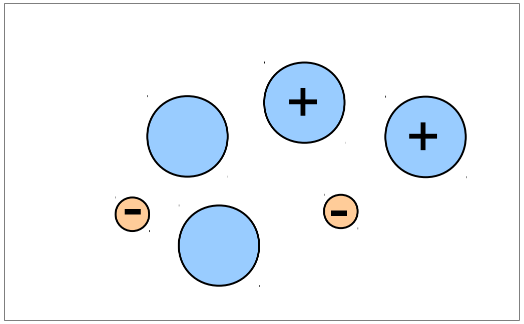

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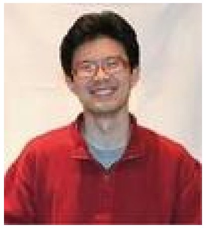

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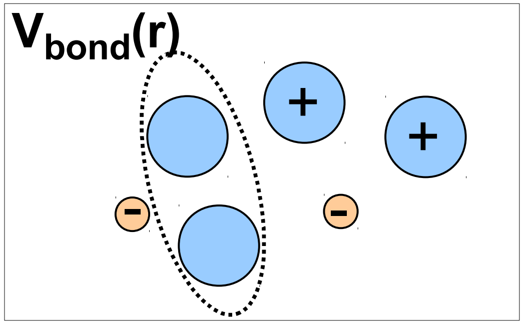

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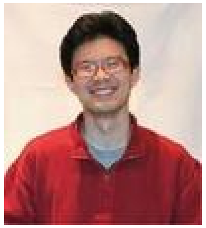

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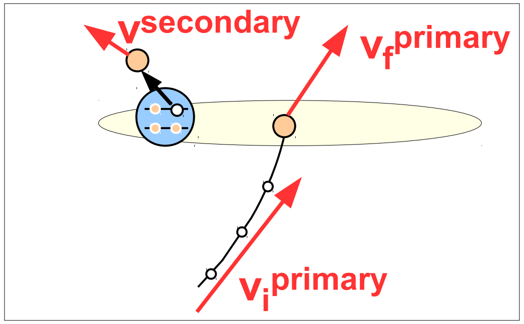

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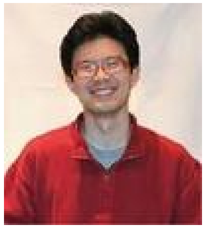

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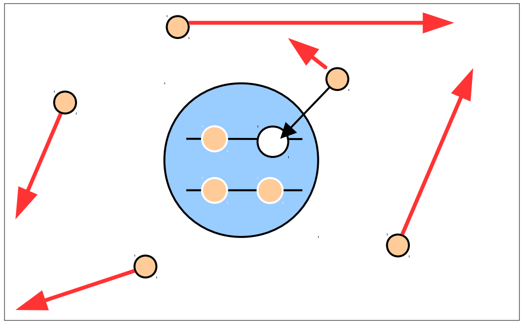

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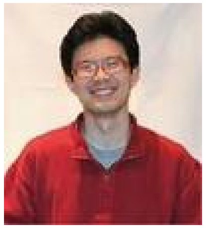

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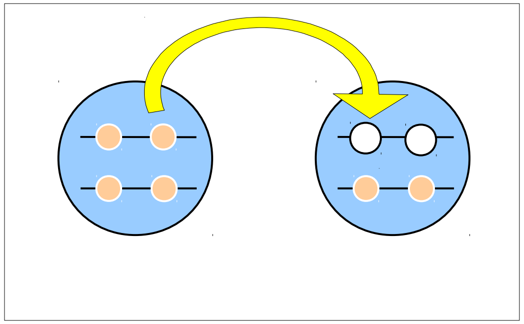

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Computational tool: XMDYN

Atomistic Particle Approach + Molecular Dynamics (MD)

One XMDYN **run** → **One realization** of the stochastic dynamics

Microscopic description
access to particle position, kinetic energy, charge state, etc.
→
Observables
distributions,
spectra
structural dynamics

Jurek, Son, Ziaja & Santra, *J. Appl. Cryst.* **49**, 1048 (2016)
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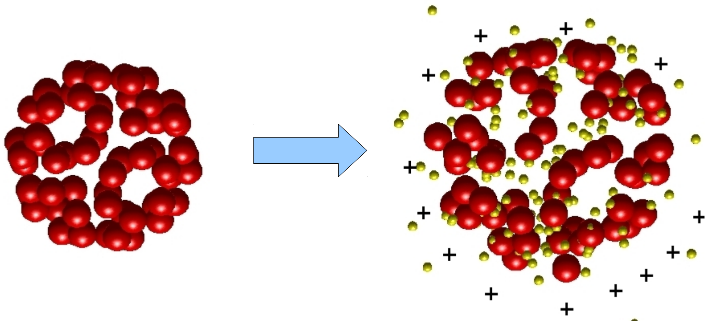
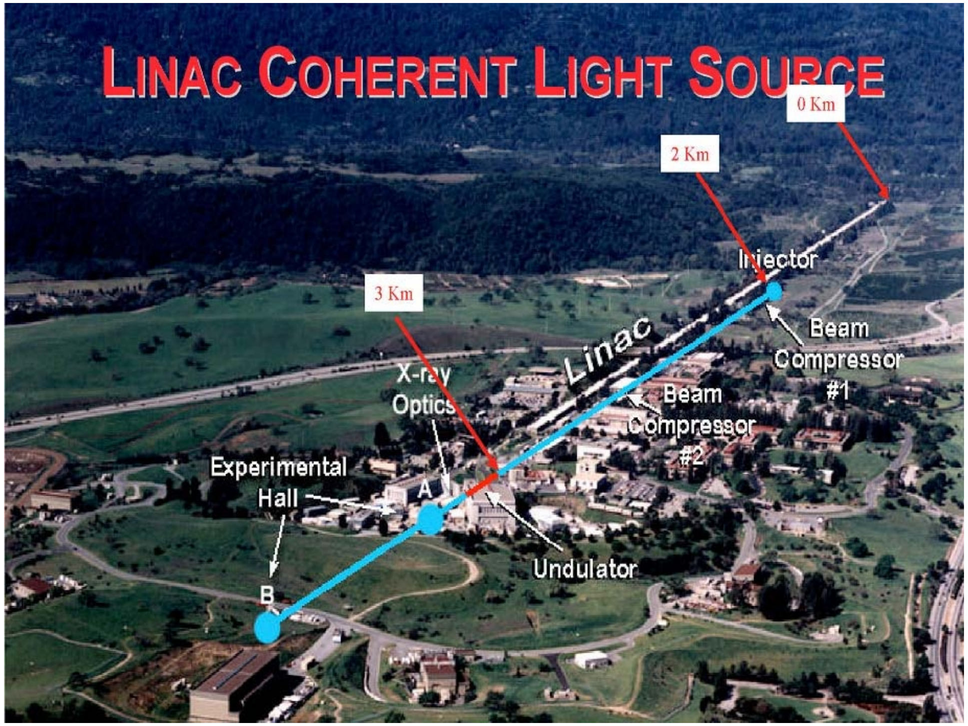
Gas phase C_{60} molecules at high x-ray intensity

C_{60} @ LCLS – The Experiment

- **Nora Berrah** (WMU) *et al.*

  C_{60} molecules irradiated **at LCLS**

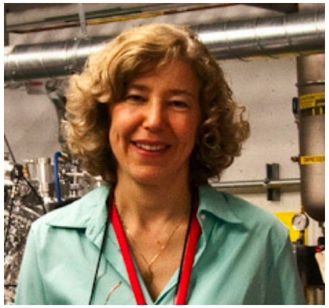




- The Goal:

  to learn about the XFEL-induced dynamics of a highly ionized complex system via **spectroscopy**

C_{60} @ LCLS – The Project & Collaboration

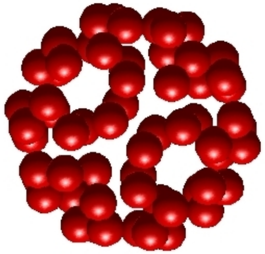


- **Experiment: Nora Berrah**

  B. F. Murphy, T. Osipov, L. Fang, M. Mucke, J.H.D. Eland, V. Zhaunerchyk, R. Feifel, L. Avaldi, P. Bolognesi, C. Bostedt, J. D. Bozek, J. Grilj, M. Guehr, L. J. Frasinski, J. Glownia, D. T. Ha, K. Hoffmann, E. Kukk, B. K. McFarland, C. Miron, E. Sistrunk, R. J. Squibb, K. Ueda

- **Theory: CFEL-DESY Theory Division**

  Z. Jurek, S.-K. Son, R. Santra



Irradiation conditions:

- $\hbar\omega$ = 485 – 800 eV
- T = 4 – 90 fs
- ε = 0.3 – 0.9 mJ

Ion data measured

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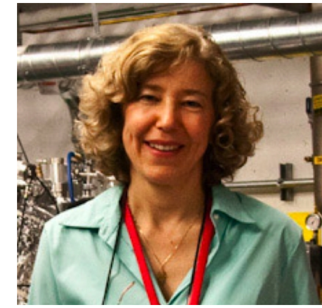
B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

C_{60} @ LCLS – The Project & Collaboration



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Irradiation conditions:

High Intensity:

Fluence ~ 15 × (1 / $\sigma^{C}_{ph.ion.}$)

ion data measured

> Theory: CFEL-DESY Theory Division

Z. Jurek, S.-K. Son, R. Santra

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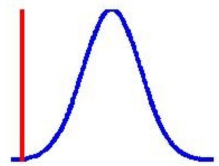
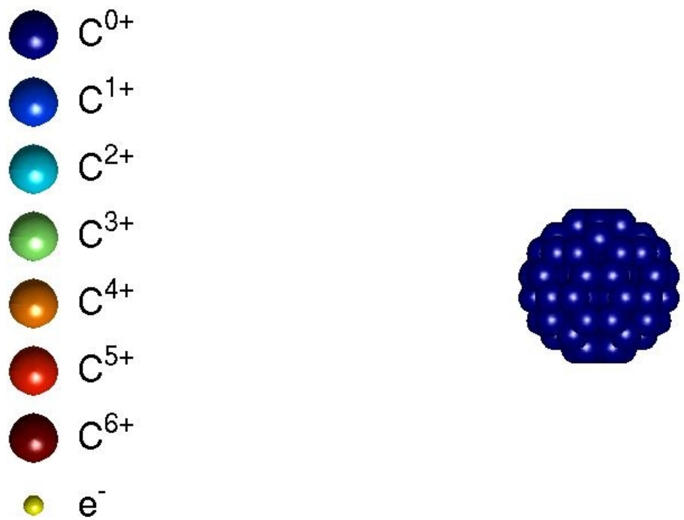
B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

> Explosion in the focus





Pulse parameters:

T = 30fs,

$\hbar\omega$ = 485eV , ε = 0.345mJ,

focus = $(1.4\mu m)^2$

double Gaussian

beam profile

-40.0fs

B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

C_{60} @ LCLS – Strong ionization (high intensity) case

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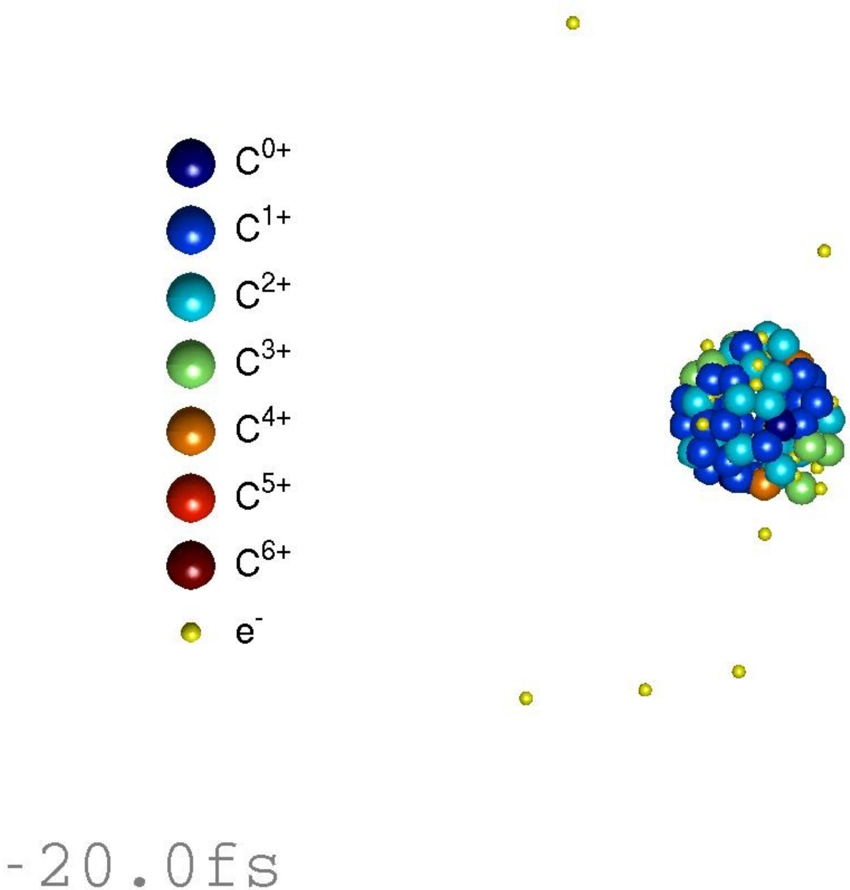

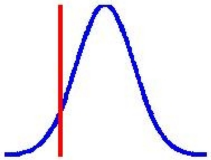

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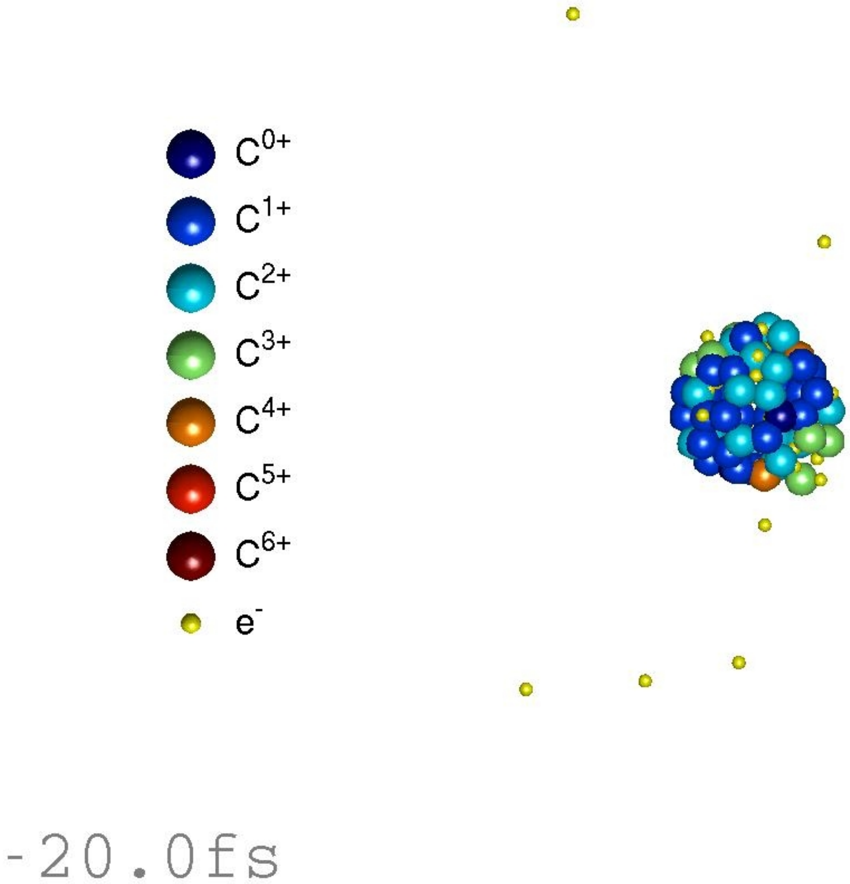

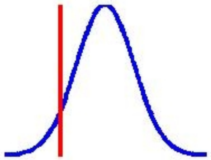

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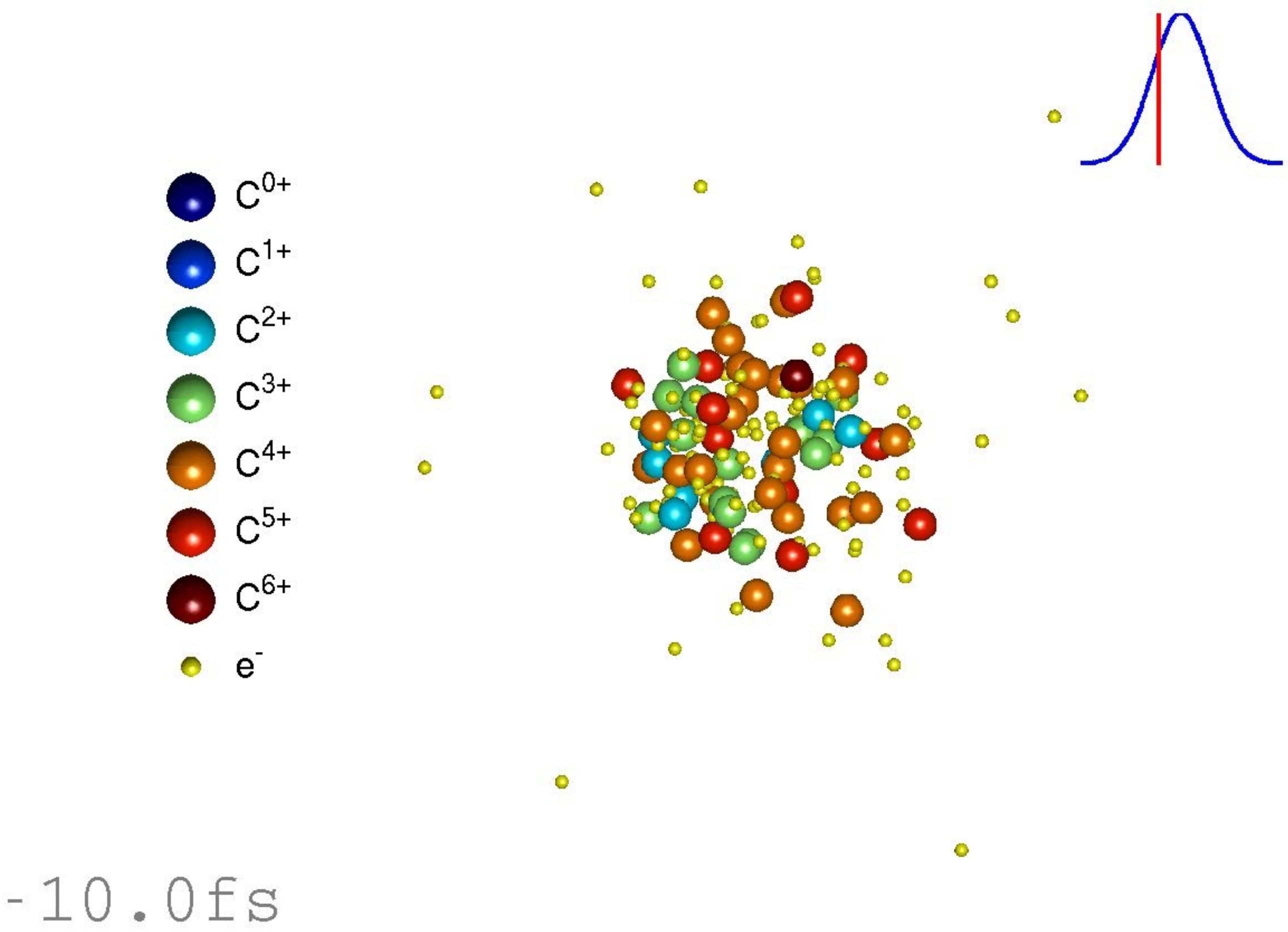

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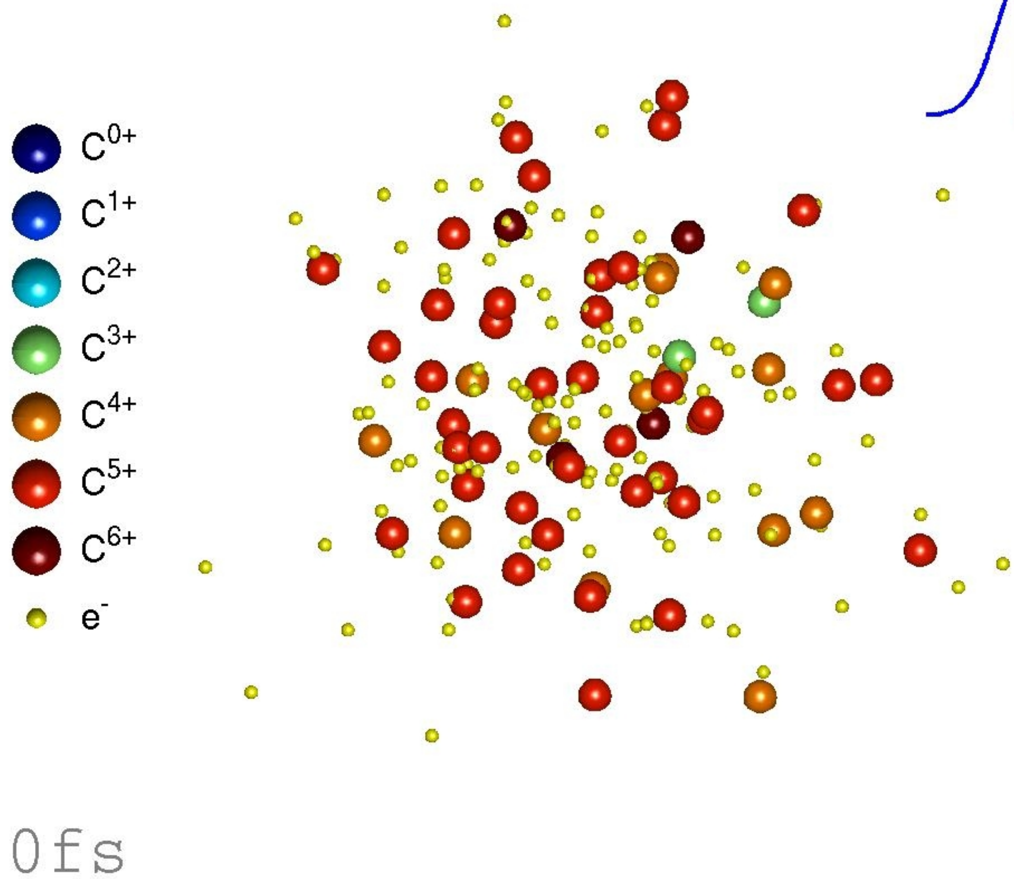

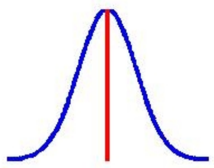

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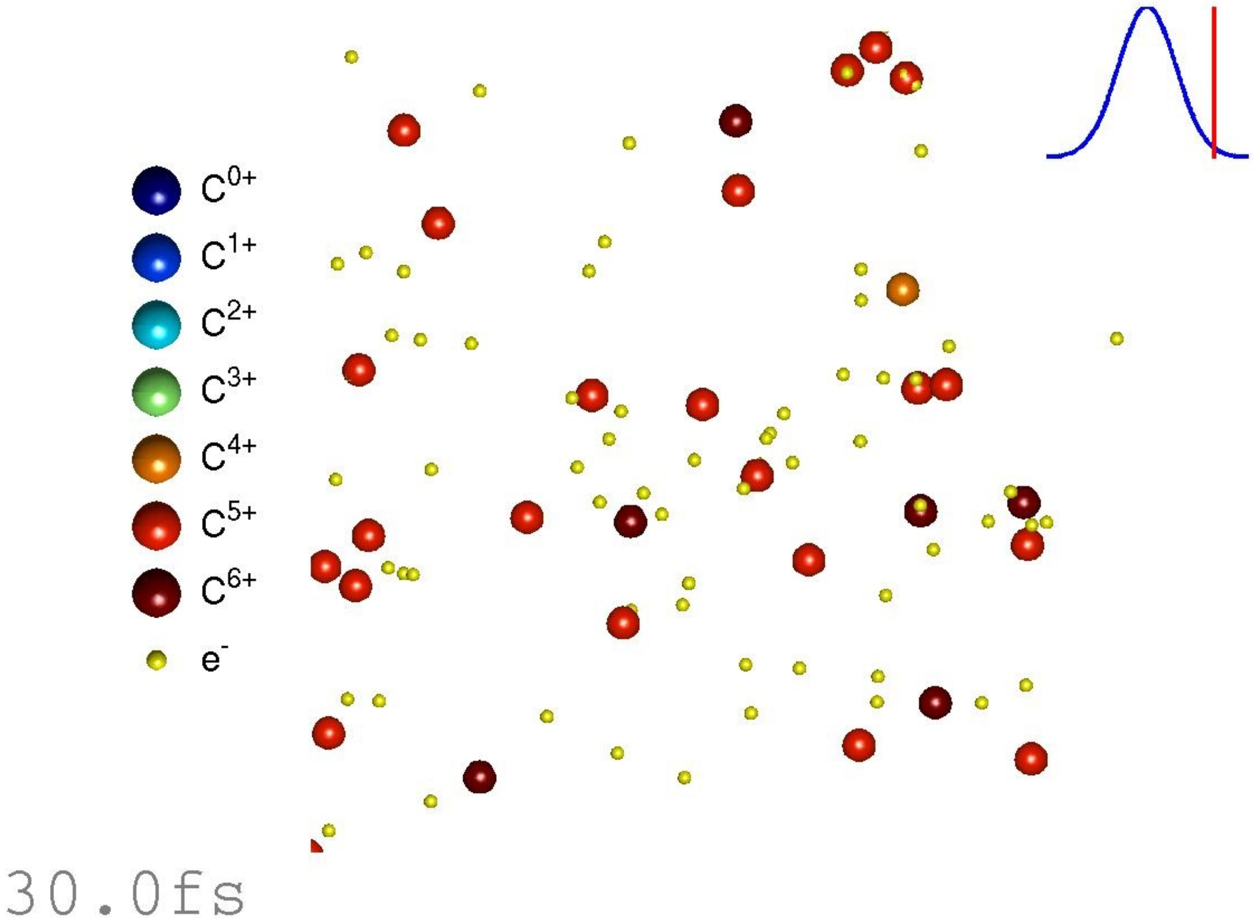

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B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

C_{60} @ LCLS – The Observables. Experiment vs. Theory

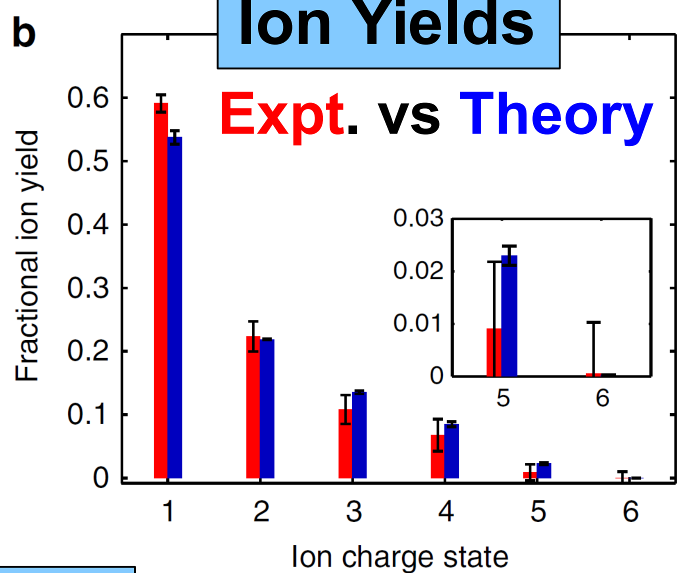
> **Atomic ions**

Theory: **No parameter fitting**!

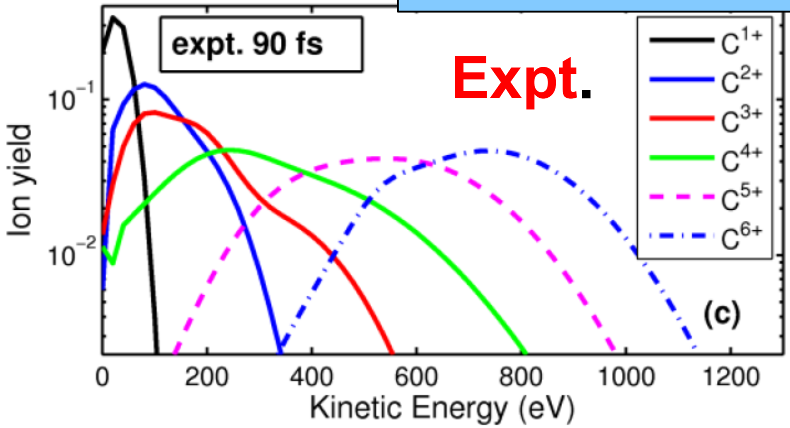
B. Murphy *et al.*, Nat. Commun. **5** 4281 (2014)

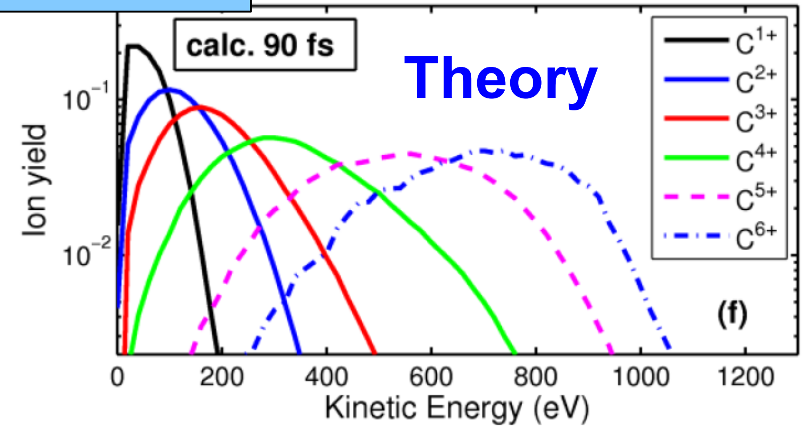
N. Berrah *et al.*, Faraday Discuss. **171** 471 (2014)

Ion Yields



Kinetic Energy spectra





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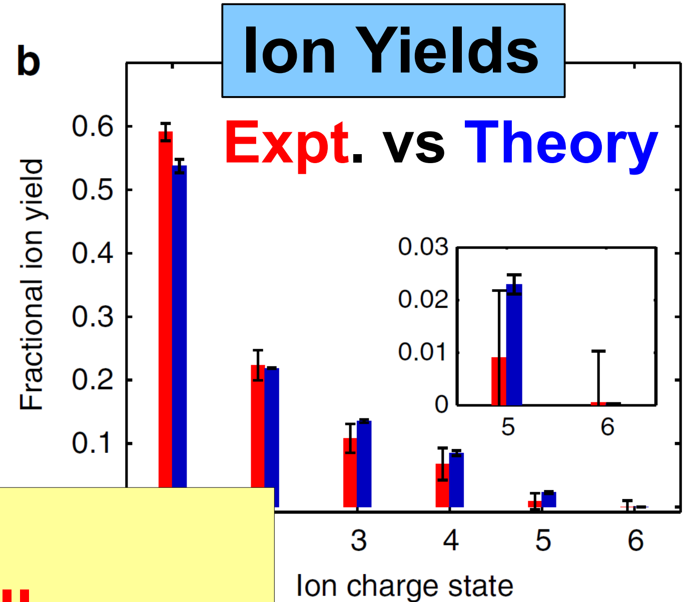
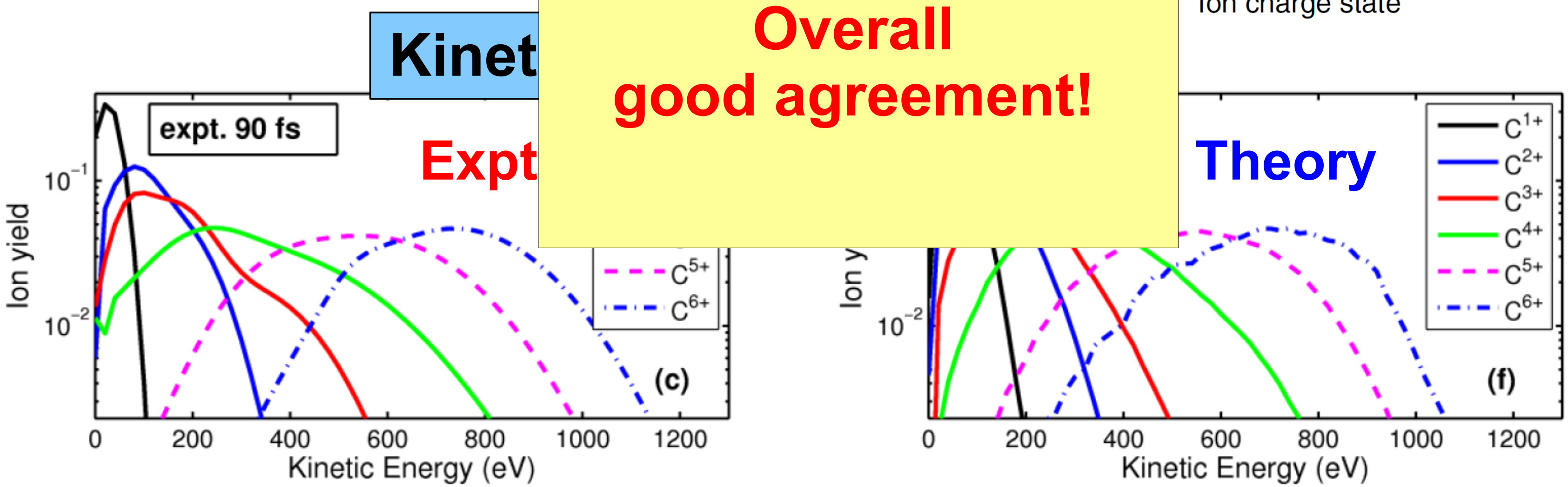




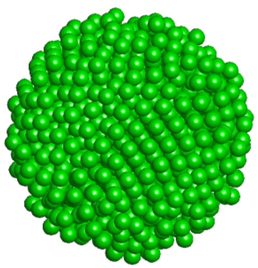
Overall good agreement!

Rare gas atomic clusters at high x-ray intensity

Rare gas clusters @ SACLA – The Experiment

- **Kiyoshi Ueda** (Tohoku Univ.) *et al.*

  Ar, Xe clusters irradiated **at SACLA**



- The Goal:

  to learn about the properties of nanoplasma formed due to XFEL irradiation via **spectroscopy**

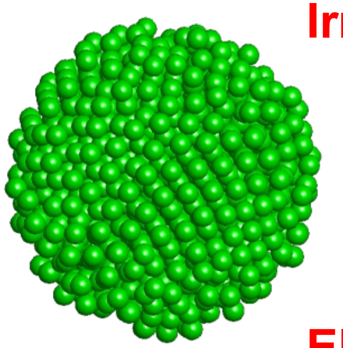
Rare gas clusters @ SACLA – The Collaboration

- **Experiment: Kiyoshi Ueda**

  T. Tachibana, H. Fukuzawa, K. Motomura, K. Nagaya, S. Wada, P. Johnsson, M. Siano, S. Mondal, Y. Ito, M. Kimura, T. Sakai, K. Matsunami, H. Hayashita, J. Kajikawa, X.-J. Liu, E. Robert, C. Miron, R. Feifel, J. P. Marangos, K. Tono, Y. Inubushi, M. Yabashi, M. Yao

- **Theory: CFEL-DESY Theory Division**

  Z. Jurek, S.-K. Son, B. Ziaja, R. Santra



Irradiation conditions:

- $\hbar\omega = 5 – 5.5$ keV
- $T = 10$ fs
- $\varepsilon \sim 0.24$ mJ

Electron data measured

T. Tachibana, Sci. Rep. **5** 10977 (2015)

Rare gas clusters @ SACLA – The Collaboration

- **Experiment: Kiyoshi Ueda**

  T. Tachibana, H. Fukuzawa, K. Motomura, K. Nagaya, S. Wada, P. Johnsson, M. Siano, S. Mondal, Y. Ito, M. Kimura, T. Sakai, K. Matsunami, H. Hayashita, J. Kajikawa, X.-J. Liu, E. Robert, C. Miron, R. Feifel, J. P. Marangos, K. Tono, Y. Inubushi, M. Yabashi, M. Yao

- **Theory: CFEL-DESY Theory Division**

  Z. Jurek, S.-K. Son, B. Ziaja, R. Santra

High Intensity:

Fluence ~ 0.16 × (1 / $\sigma^{Ar}_{ph.ion.}$)

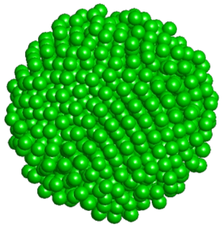
T. Tachibana, Sci. Rep. **5** 10977 (2015)

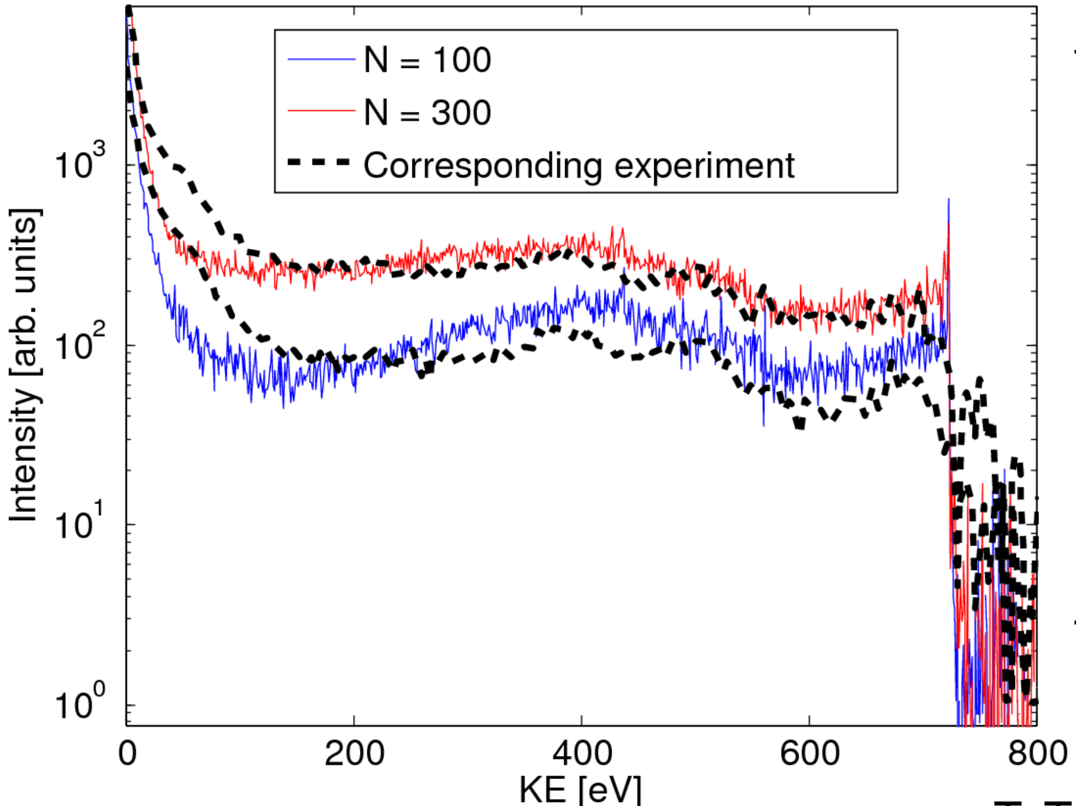
Application 2: Clusters @ SACLA

> **Theoretical and experimental electron kinetic energy spectra**,

ħω = 5.5keV, T=10fs

Xe_{100}, Xe_{300}





– Challenge for Theory:

23.532.201 possible electronic configurations / atom

XATOM **on-the-fly** XMDYN

– Theory:

No parameter fitting!

T. Tachibana, Sci. Rep. **5** 10977 (2015)

Application 2: Clusters @ SACLA

> **Theoretical and experimental electron kinetic energy spectra**,

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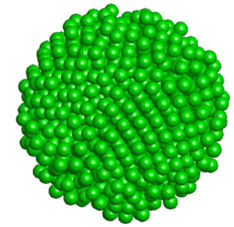
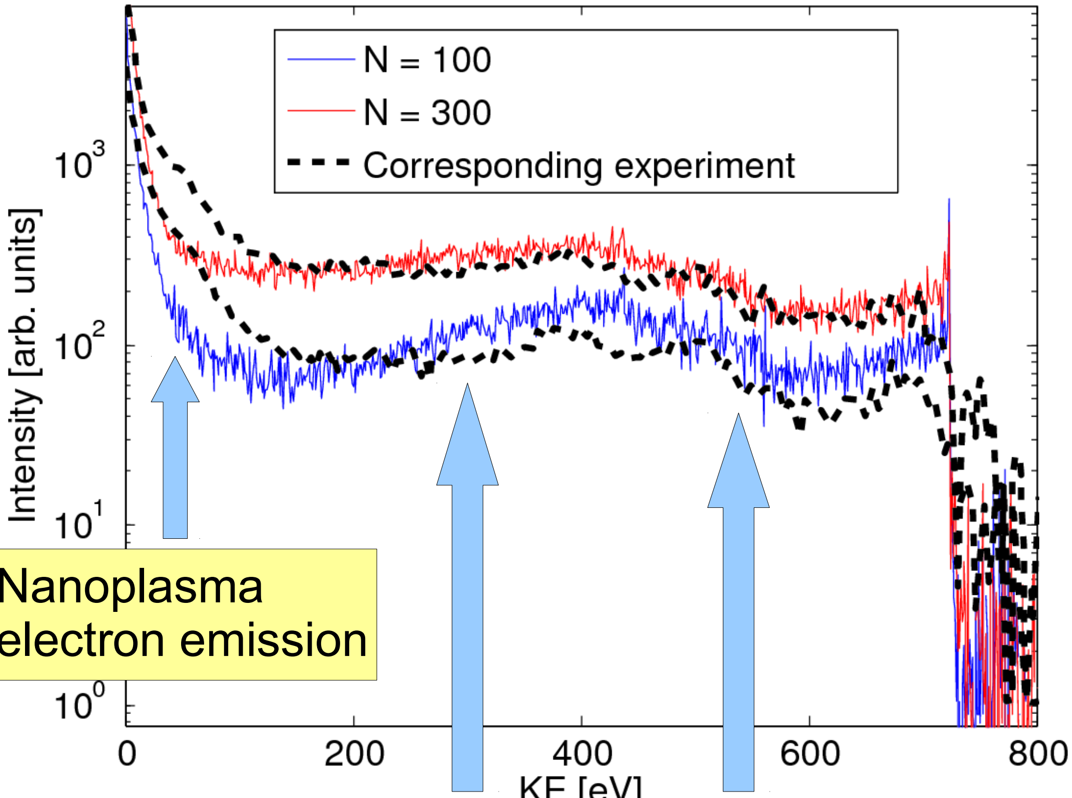




Nanoplasma electron emission

Slowed down Photo & Auger electrons

– Challenge for Theory:

23.532.201 possible electronic configurations / atom

XATOM $\xrightarrow{\text{on-the-fly}}$ XMDYN

– Theory:

No parameter fitting!

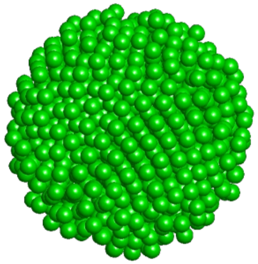
T. Tachibana, Sci. Rep. **5** 10977 (2015)

Rare gas cluster dynamics
at moderate x-ray intensities:
Chemical effects

Rare gas clusters @ SACLA – The Experiment

- **Kiyoshi Ueda** (Tohoku Univ.) *et al.*

  Ar_{1000} clusters irradiated **at SACLA**



- The Goal: to learn about the **fragmentation dynamics** due to multiple ionization via **spectroscopy**

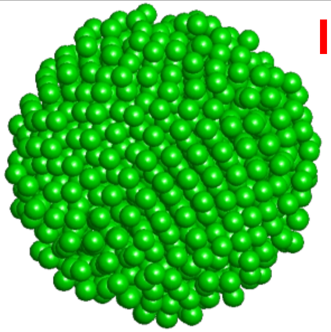
Rare gas clusters @ SACLA – The Collaboration

> **Experiment: Kiyoshi Ueda**

Y. Kumagai, W. Xu, H. Fukuzawa, K. Motomura, D. Iablonskyi, K. Nagaya, S.-i. Wada, S. Mondal,T. Tachibana, Y. Ito, T. Sakai, K. Matsunami, T. Nishiyama, T. Umemoto, C. Nicolas, C. Miron, T. Togashi, K. Ogawa, S. Owada, K. Tono, M. Yabashi

> **Theory: CFEL-DESY Theory Division**

Z. Jurek, S.-K. Son, B. Ziaja, R. Santra



Irradiation conditions:

– $\hbar\omega$ = 5.5 keV

– T = 10 fs

– Fluence ~ 4 $\mu J/\mu m^2$

Ion and electron data measured

Y. Kumagai *et al*, *submitted*

Rare gas clusters @ SACLA – The Collaboration

- **Experiment: Kiyoshi Ueda**

  Y. Kumagai, W. Xu, H. Fukuzawa, K. Motomura, D. Iablonskyi, K. Nagaya, S.-i. Wada, S. Mondal,T. Tachibana, Y. Ito, T. Sakai, K. Matsunami, T. Nishiyama, T. Umemoto, C. Nicolas, C. Miron, T. Togashi, K. Ogawa, S. Owada, K. Tono, M. Yabashi

- **Theory: CFEL-DESY Theory Division**

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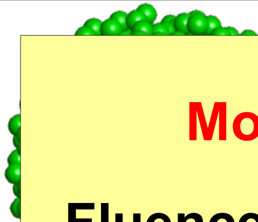


Moderate Intensity:

Fluence ~ 0.01 × (1 / $\sigma^{Ar}_{ph.ion.}$)

ion and electron data measured

Y. Kumagai *et al*, *submitted*

Ar clusters at moderate x-ray intensity

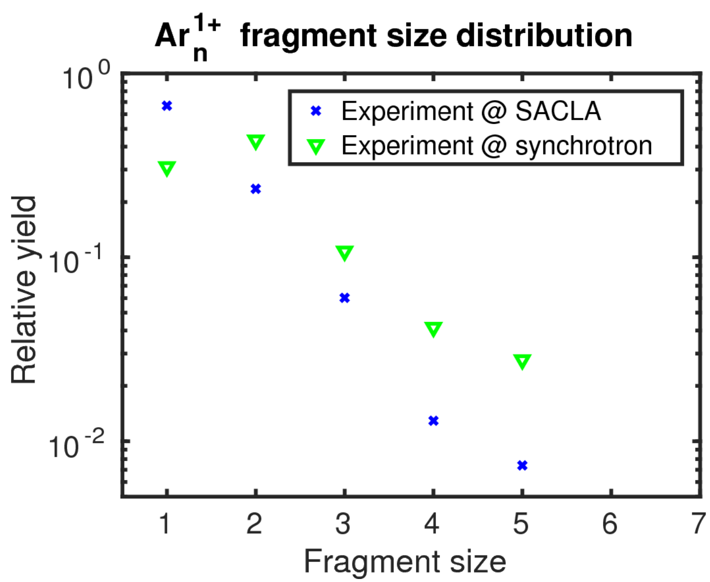
- Moderate x-ray intensity → low photoionization density

  → **low** level of **excitation**, significant role of **chemical effects**

- Ar_{1000} clusters: - at synchrotron (→ single photoionization event)

  H. Murakami *et al*, J.Chem.Phys **126** 054306 (2007)

  - at moderate XFEL intensities



- **Oligomer formation**
- Higher excitation

  → more fragmentation

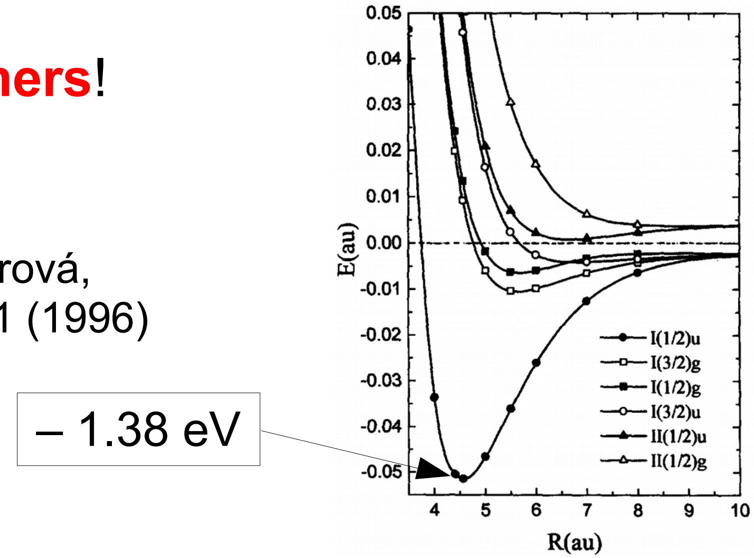
Y. Kumagai *et al*, *submitted*

Chemistry or singly charged Ar clusters

- **Chemical bond** formation in **Ar_2^{1+} dimers**!

Potential curves

F. X. Gaeda, I. Paidarová,
Chem. Phys. **209** 281 (1996)

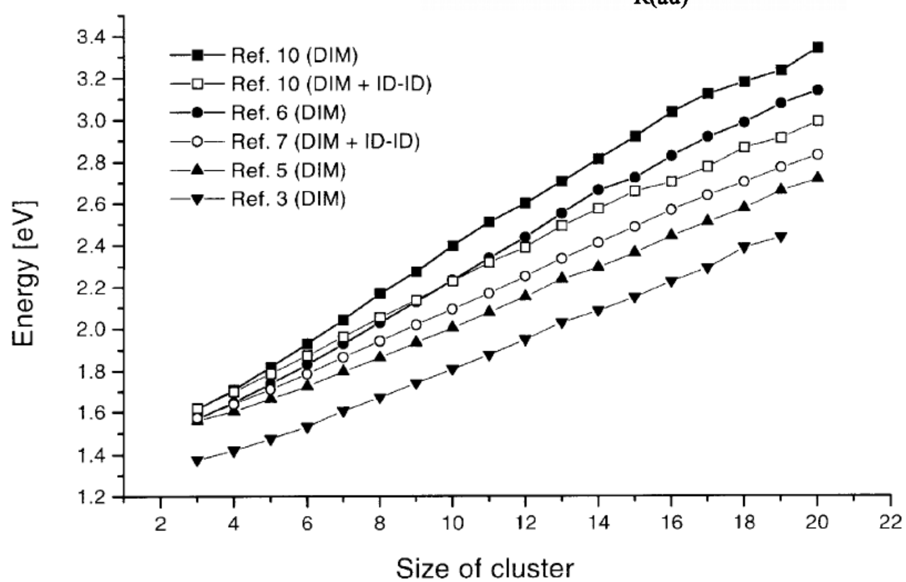


- Bonding in **Ar_n^{1+} clusters**

Dissociation energies

$$Ar_n^{1+} \rightarrow Ar^{1+} + (n-1)\,Ar$$

D. Hrivnak, R. Kalus,
Chem. Phys. **264** 319 (2001)



Model of the chemical effects (extension to XMDYN)

> **van-der-Waals** interaction

> **Chemical bond** formation

– between an ionized and neutral Ar sites (classical force fields)

→ strong dimer between Ar^{1+} and one neutral Ar neighbor

→ weak interaction between Ar^{1+} and other neutral atoms

– bond switching at potential curve crossings

→ dimer formation and disappearance during the dynamics

> **charge transfer** between sites: > over-the-barrier approach

> valence orbitals involved

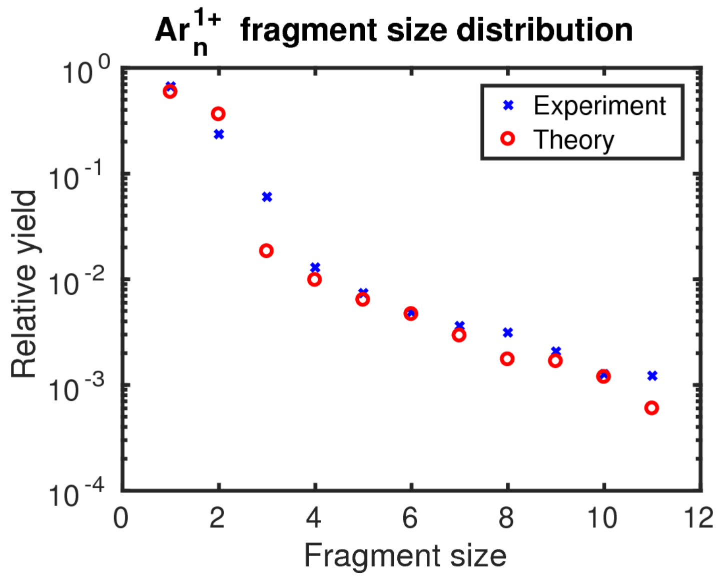
Y. Kumagai *et al*, *submitted*

Ar clusters at moderate x-ray intensity

- Moderate x-ray intensity → low photoionization density

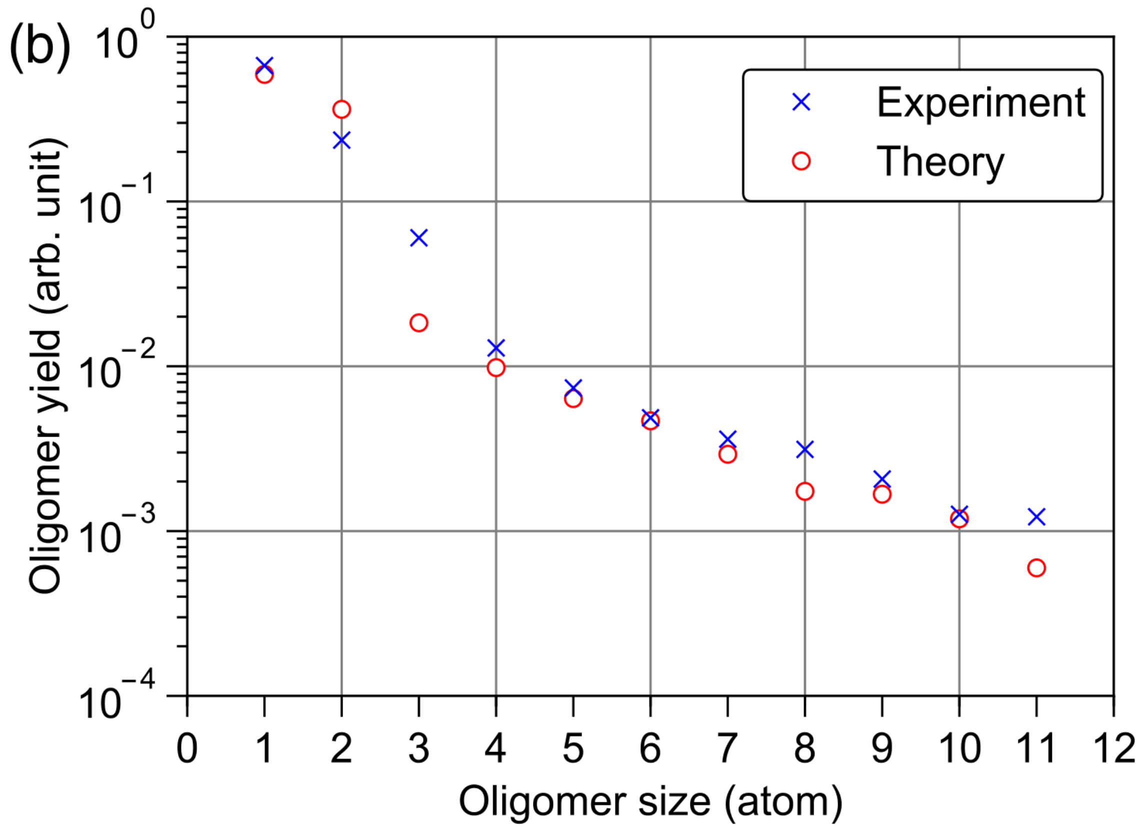
  → low level of excitation, significant role of **chemical effects**

- Ar_{1000} clusters fragment yield: **Experiment vs. Theory**



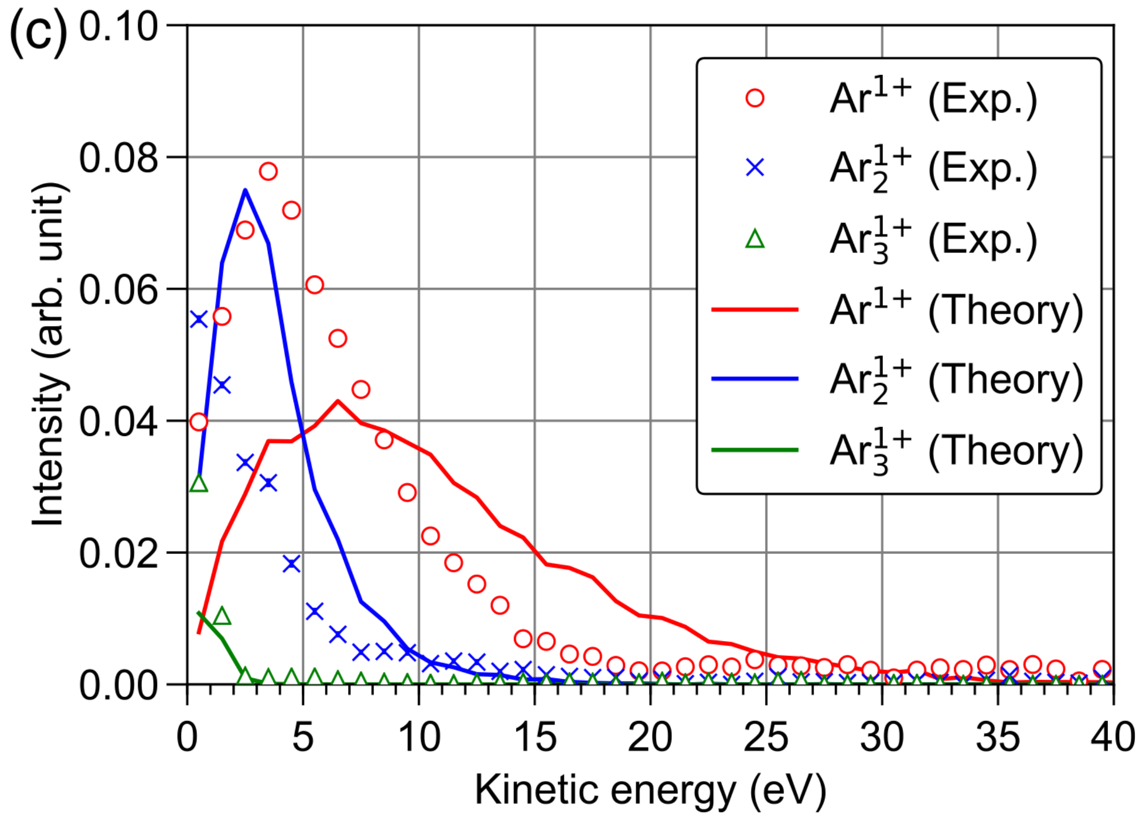
Y. Kumagai *et al*, PRL **120** 223201 (2018)

Oligomer yield



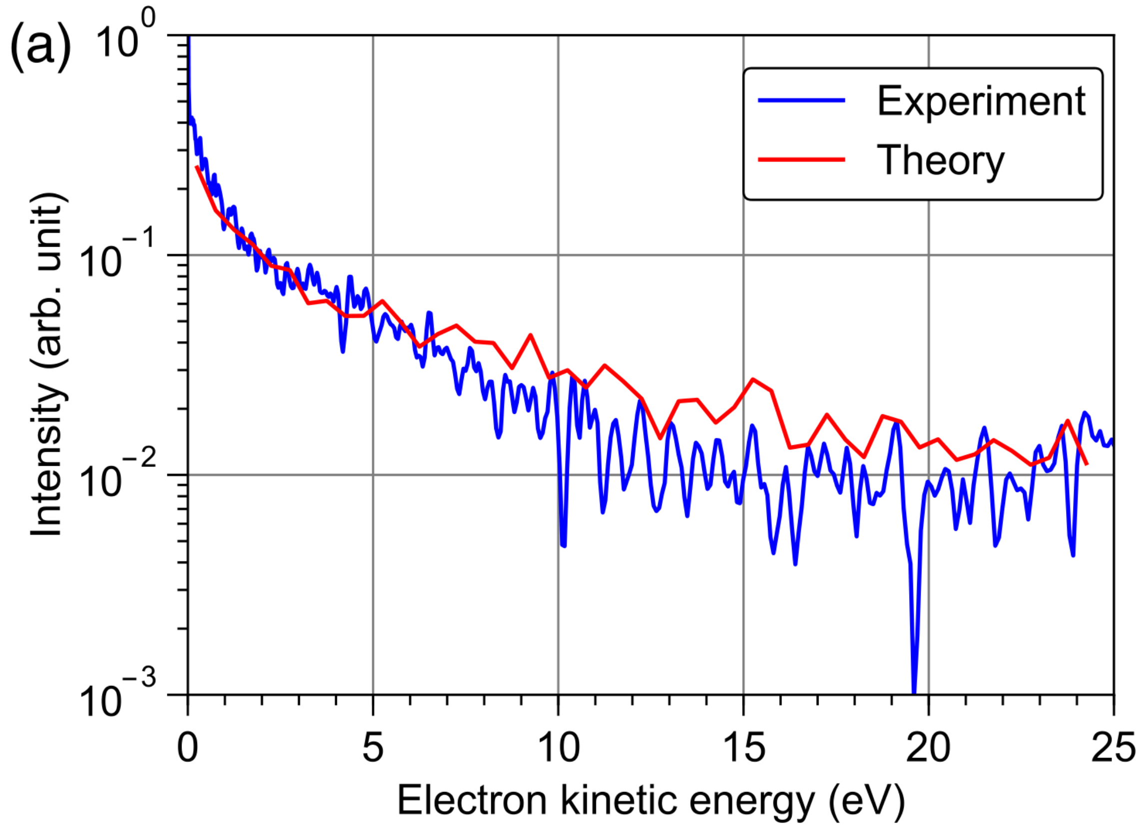
Y. Kumagai *et al*, PRL **120** 223201 (2018)

Kinetic energy spectra of oligomers



Y. Kumagai *et al*, PRL **120** 223201 (2018)

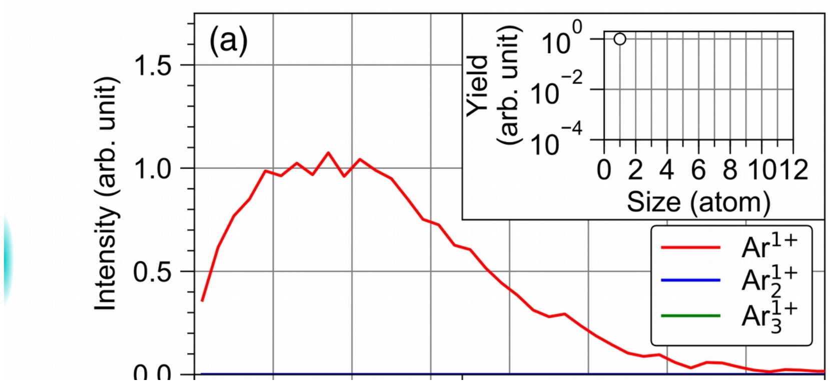
Kinetic energy spectrum of electrons



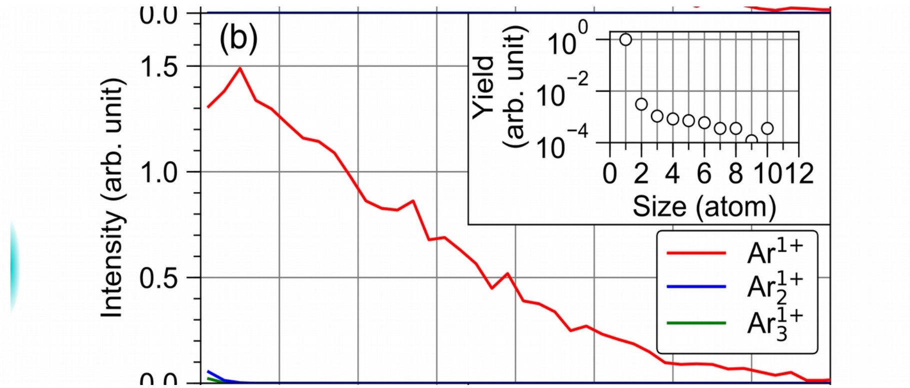
Y. Kumagai *et al*, PRL **120** 223201 (2018)

Predictions with model variants for fixed fluence – ion KE

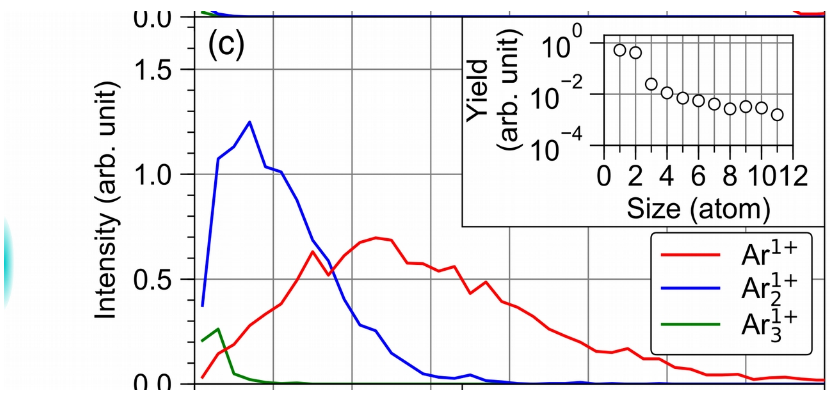
No chemical effect



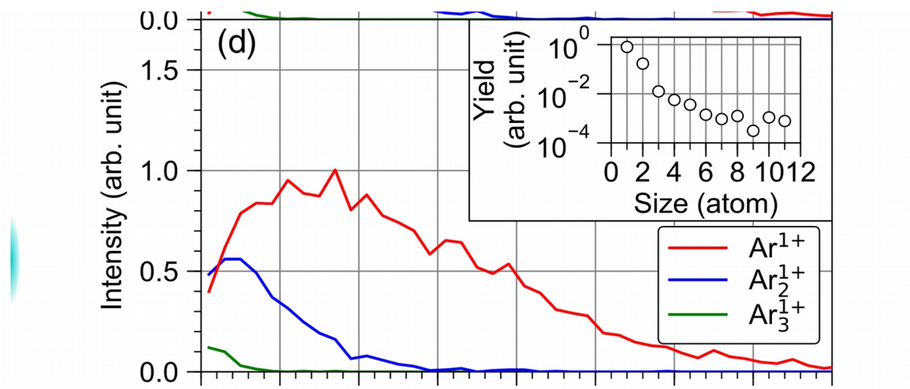
van de Waals added



van der Waals + dimer formation



van der Waals + dimer + charge transfer



Y. Kumagai *et al*, PRL **120** 223201 (2018)

Summary

- Molecular Dynamics based **modeling framework**

  XMDYN, XATOM

  Heavy atoms

- **Clusters, molecules** at high x-ray intensity

  – strongly bound fullerens

  – rare gas cluster and **chemical effects**

- **Perspectives** for Simulations

  – **Bulk** system (extreme states of matter, nanocrystallography)

  – **Complex simulation** framework (S2E, SIMEX)