Ab-initio calculation of electron impact ionization cross sections for ions in exotic electron configurations

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Single Particle Imaging

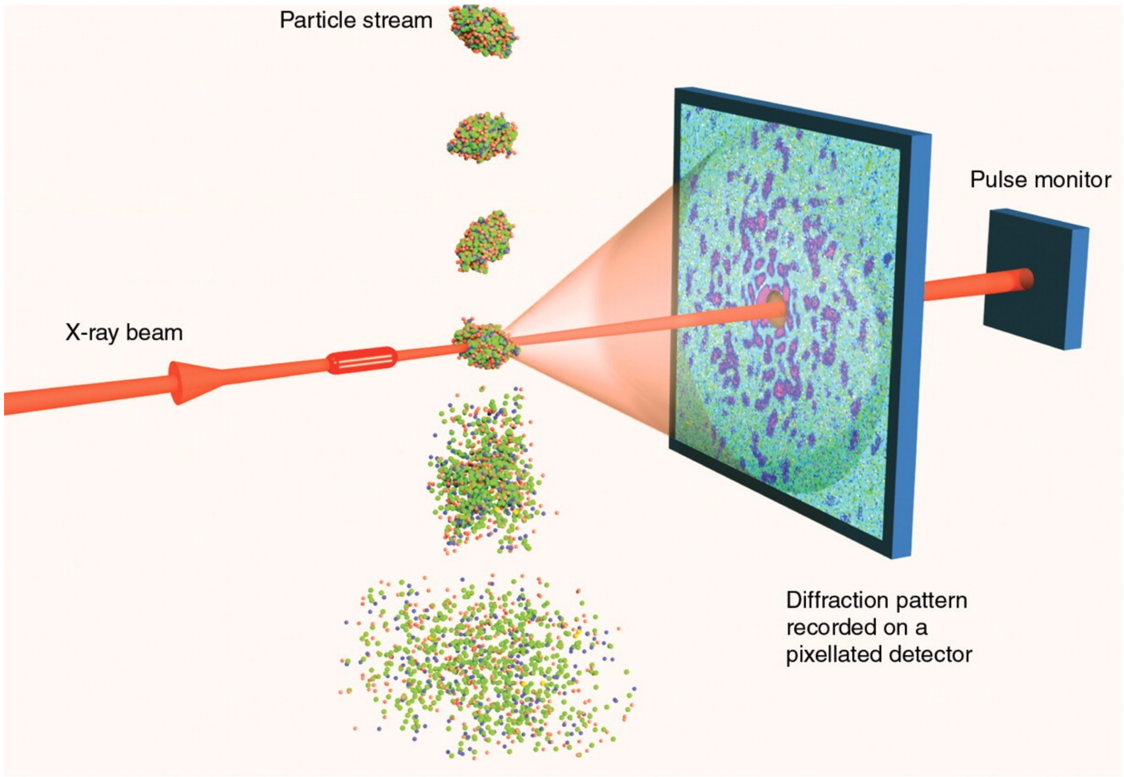
- X-ray Free Electron Lasers (XFELs):

  ultrashort pulses of hard x-rays with high fluence

  Duration ~ 1 fs
  Energy ~ 250 eV - 25 keV
  Fluence ~ 10^{12} - 10^{13} photons/pulse

- X-ray crystallography and single particle imaging

- Diffraction before destruction: pulse induces radiation damage

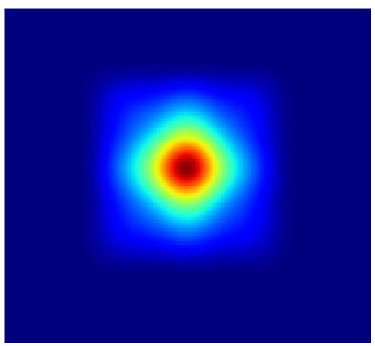


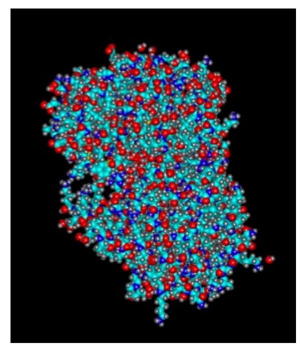
K.J. Gaffney, H.N. Chapman, Science **316**, 1444-1448 (2007)

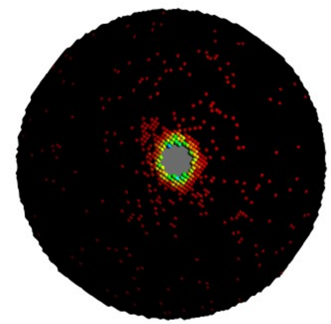
Start-to-End Simulations

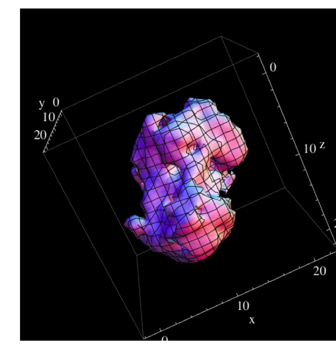
http://www.xfel.eu/sims2e

FEL source → Optics → Photon/Matter Interaction → Diffr. Patterns → Analysis









Images: courtesy of A. Mancuso and collaborators

Yoon et al, Sci. Rep. **6**, 24791 (2016)

XMDYN

Microscopic simulation of photo-induced dynamics

Jurek et al, J. Appl. Crystallogr., **49**, 1048-1056 (2016)

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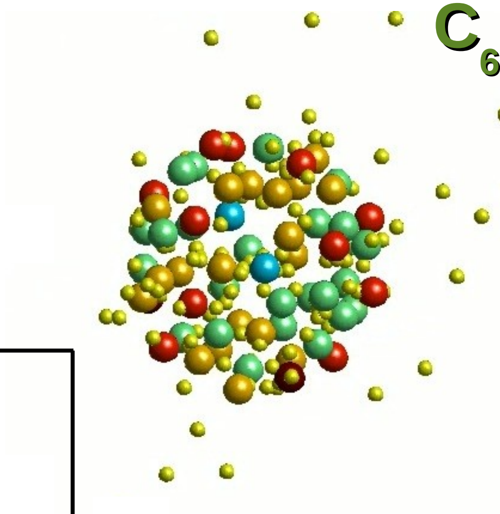
XMDYN

- Atomistic Model + Molecular Dynamics

C_{60}



Atomistic Model	Molecular Dynamics
• Inner-shell processes incorporated • Rates and cross sections provided by XATOM[1]	• Atoms/ions and electrons treated classically • Dynamics through classical force fields

- Environmental effects also considered
- Many XMDYN runs for Monte-Carlo simulation

[1] S.-K. Son, J.J. Bekx, K. Toyota, O. Geffert, J.M. Slowik, and R. Santra, *xatom*---an integrated toolkit for x-ray and atomic physics, CFEL, DESY, Hamburg, Germany, 2017, Rev. 2928.

XATOM

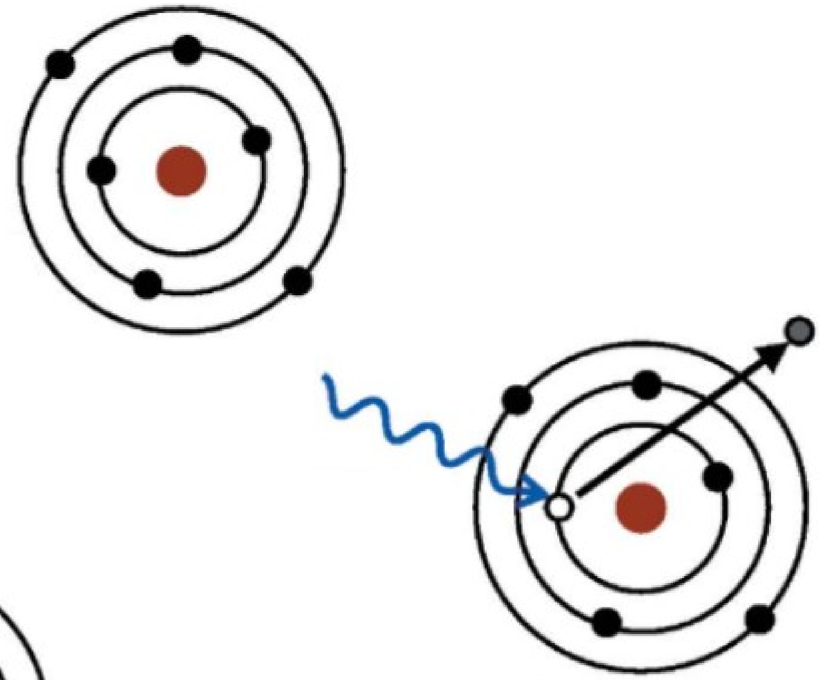
- Ab-initio toolkit for x-ray-matter interactions
- Rates and cross sections of photo-ionization, Auger decay, fluorescence and Compton scattering
- Hartree-Fock-Slater:

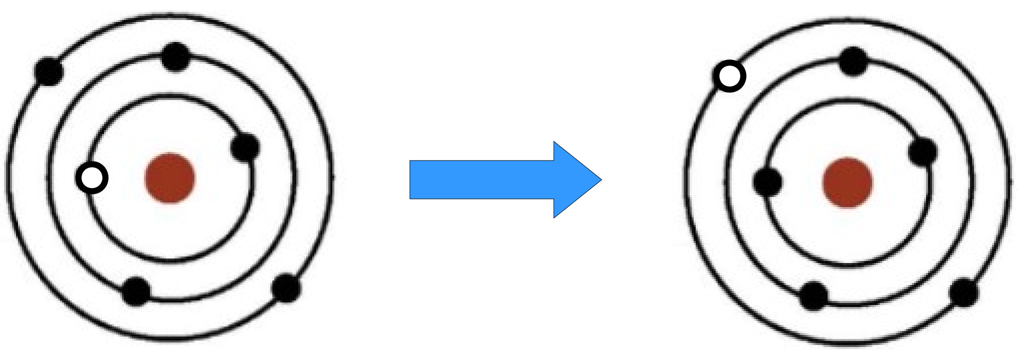
$$H_{HFS}\psi(\boldsymbol{x})=\epsilon\psi(\boldsymbol{x}), with$$

$$H_{HFS}=-\frac{1}{2}\nabla^2-\frac{Z}{|\boldsymbol{x}|}+\int d^3x'\frac{n(\boldsymbol{x}')}{|\boldsymbol{x}-\boldsymbol{x}'|}-\frac{3}{2}\left[\frac{3}{\pi}n(\boldsymbol{x})\right]^{1/3}$$

Radiation Damage

- X-ray pulses induce photo-ionization yielding core-hole ions
- Life-time of core-hole states ~1-10 fs

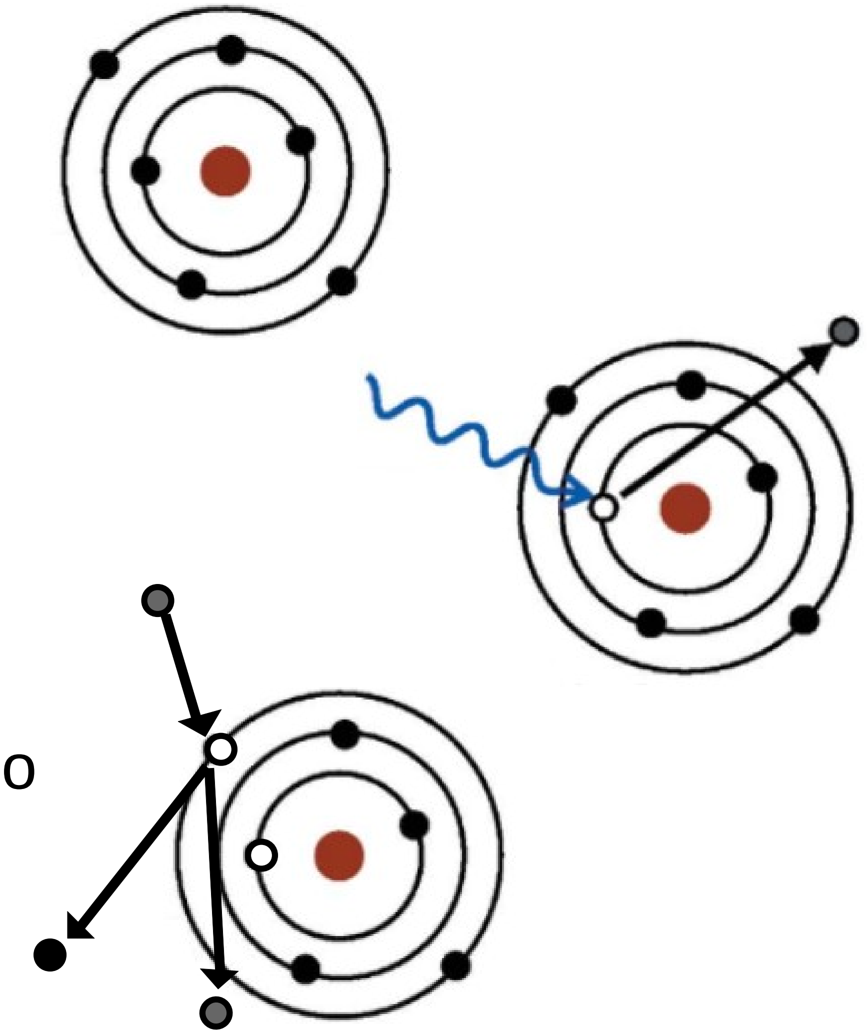




Ground-state ion

Radiation Damage

- X-ray pulses induce photo-ionization yielding core-hole ions
- Life-time of core-hole states ~1-10 fs
- Ions can undergo impact ionization with secondary electrons
- Core-hole ions can also undergo impact ionization



Doubly-Differential Electron Impact Ionization Cross Section

K. Sturm, Z. Naturforsch. **48A**, 233-242 (1993)

$$\frac{d^2\sigma}{d\Omega_{out}\, dE_{out}} = \frac{4}{Q^4}\frac{q_{out}}{q_{in}}\sum_{I,F} P_I \left|\langle F|e^{i\mathbf{Q}\mathbf{x}}|I\rangle\right|^2 \delta\left(E_{in}-E_{out}-E_F+E_I\right)$$

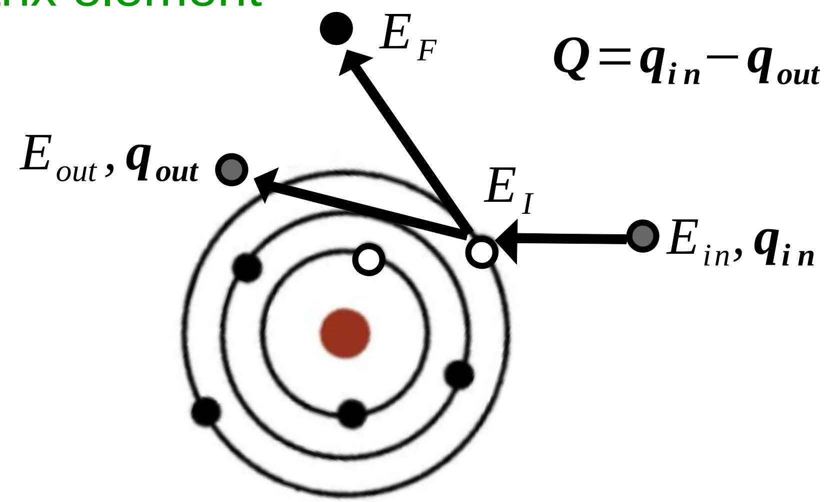
Coulomb repulsion

Statistics

Transition matrix element

Energy conservation

- Incoming and scattered electron approximated by plane waves
- Free states are approximated with a pseudo-continuum
- Implemented the above formula in the XATOM framework



Total Cross Section

- Comparison made with
  - Experiment [1,2]

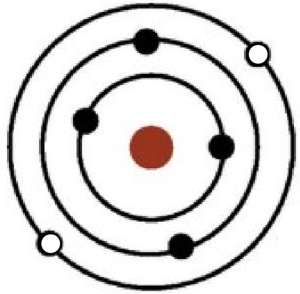
    Data only available for ground-state ions
  - Lotz formula [3] (semi-empirical)

    Best-fit approximation from available data (1970)
  - BEB method [4] (currently in use in XMDYN)

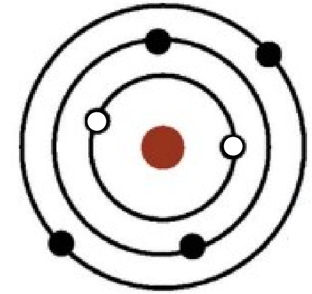
    Heuristic matching of asymptotic formulae

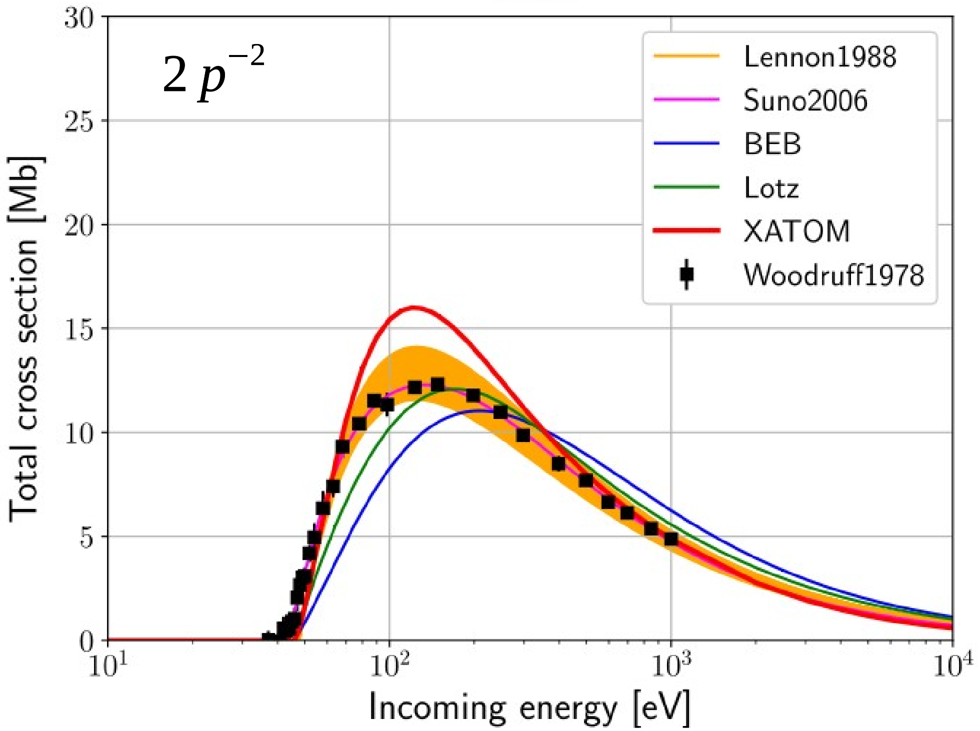
[1] M.A. Lennon et al., J. Phys. Chem. Ref. Data **17**, 1285 (1986)
[2] H. Suno and T. Kato, At. Data Nucl. Data Tables, **92** 407 (2006)
[3] W. Lotz, Z. für Phys. **232**, 101-107 (1970)
[4] Y.-K. Kim and M. E. Rudd, Phys. Rev. A **50**, 3954-3967 (1994)

Total Cross Section

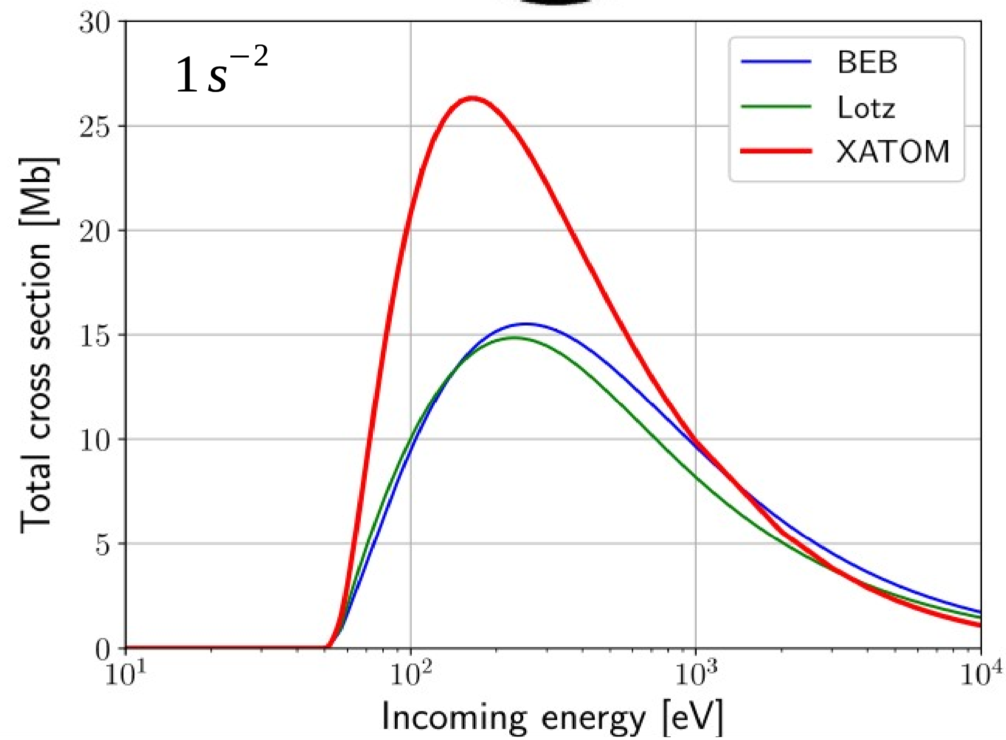


Carbon(2+) ion





Valence holes

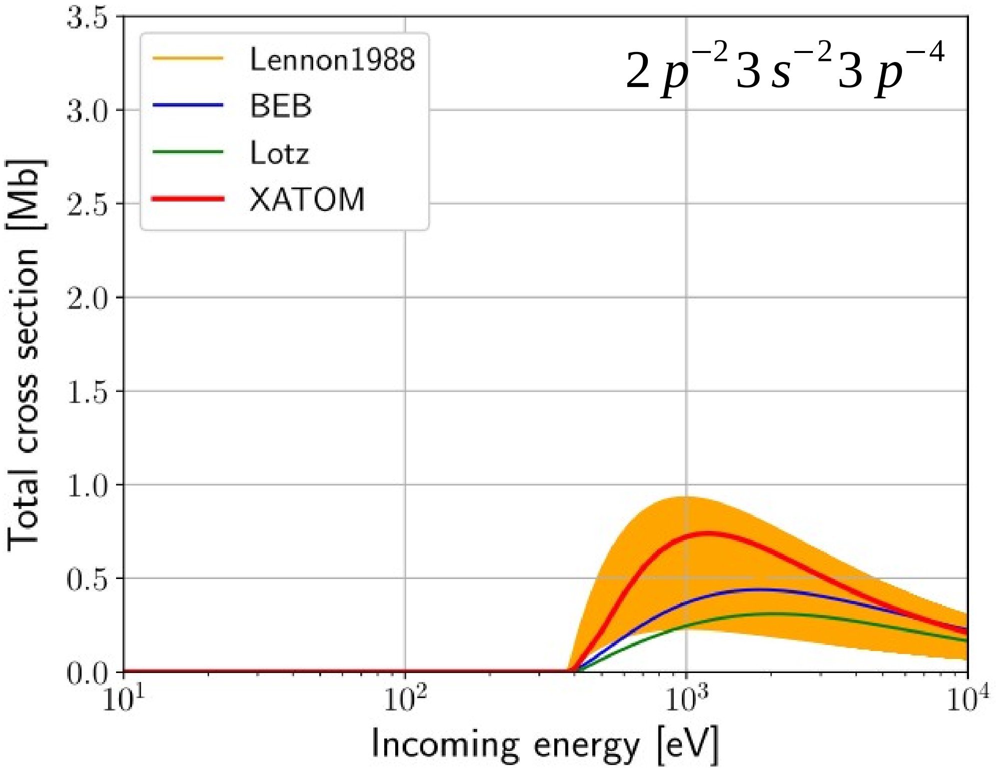


Core holes

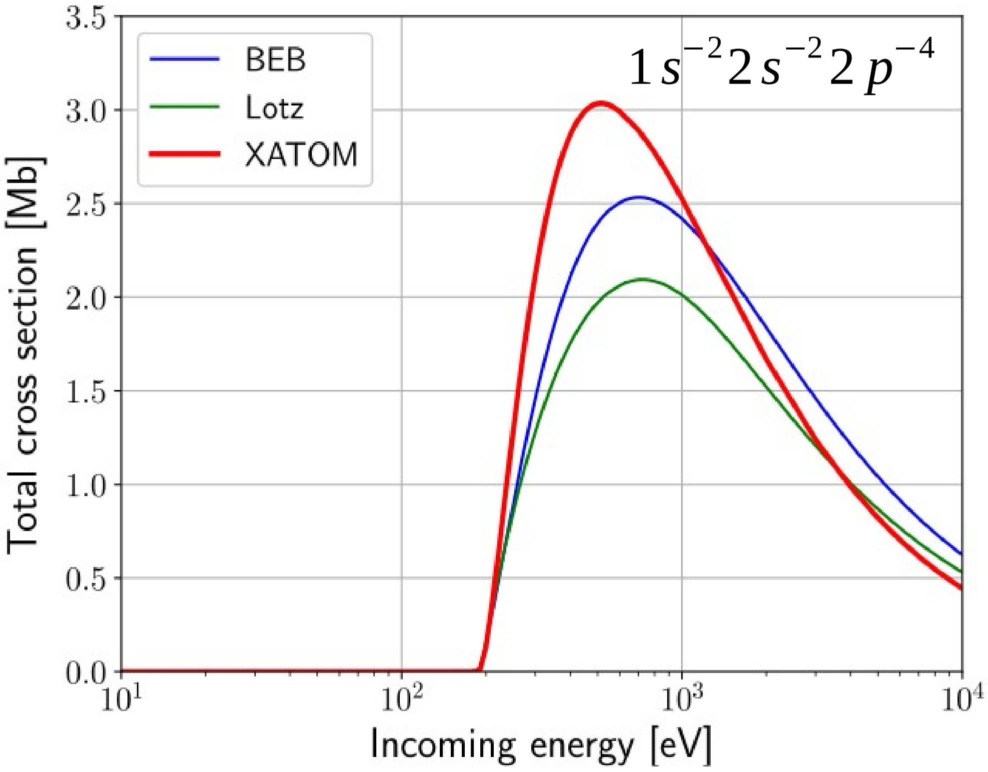
J.J. Bekx, S.-K. Son, R. Santra and B. Ziaja *(submitted)*

Total Cross Section

Sulphur(8+) ion



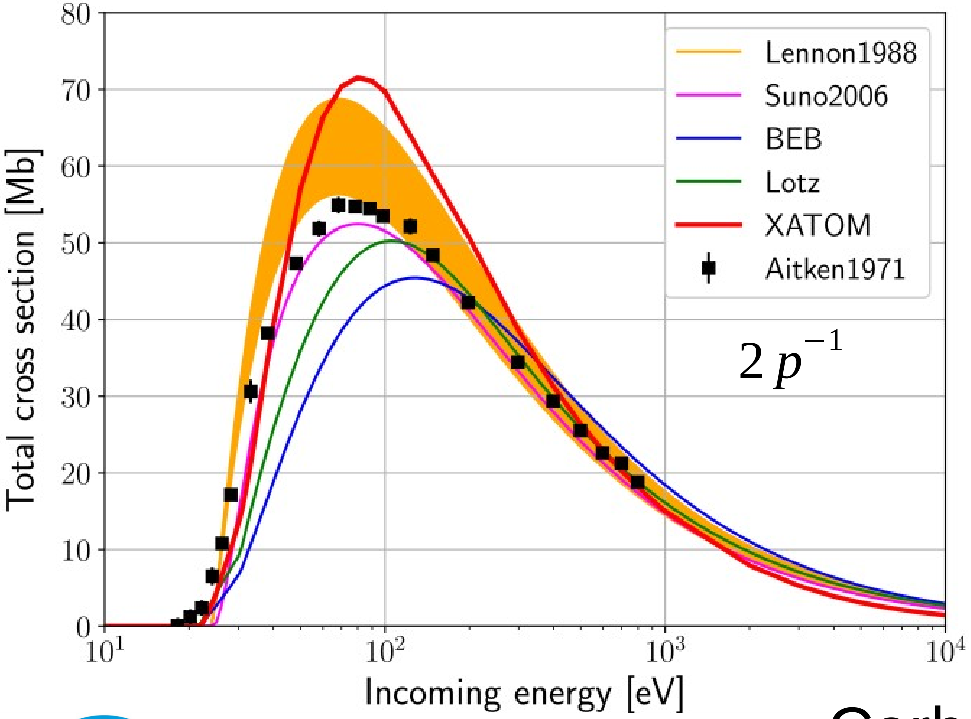
Valence holes

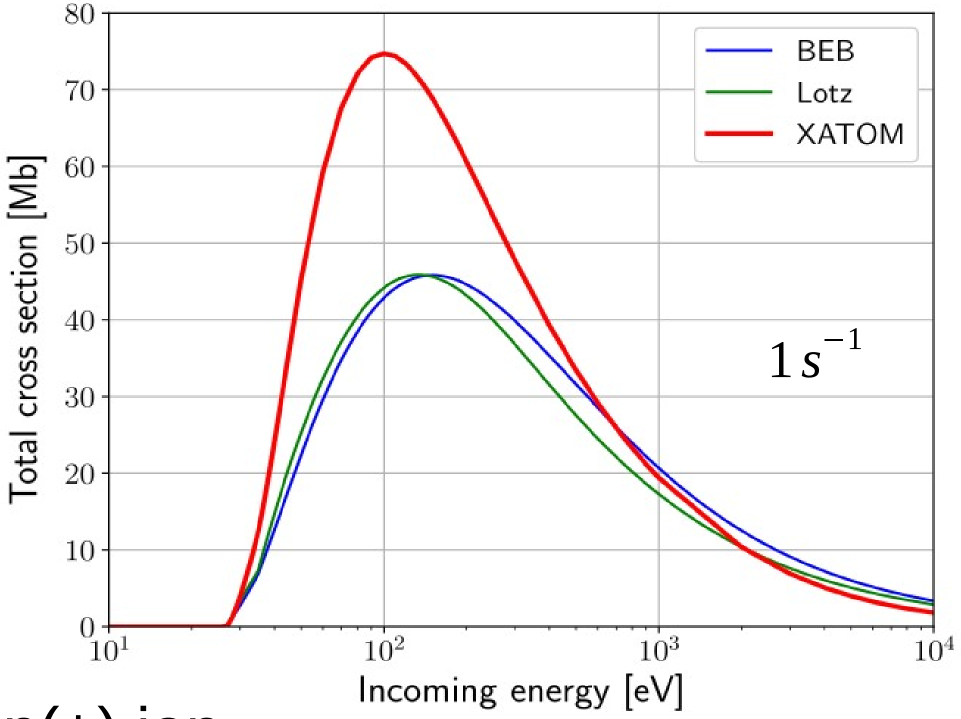


Core holes

Observations

- Total cross section results are larger for core-hole ions than for ground-state ions
- BEB mimics this behavior predicted by XATOM, but Lotz generally does not





Carbon(+) ion

Observations

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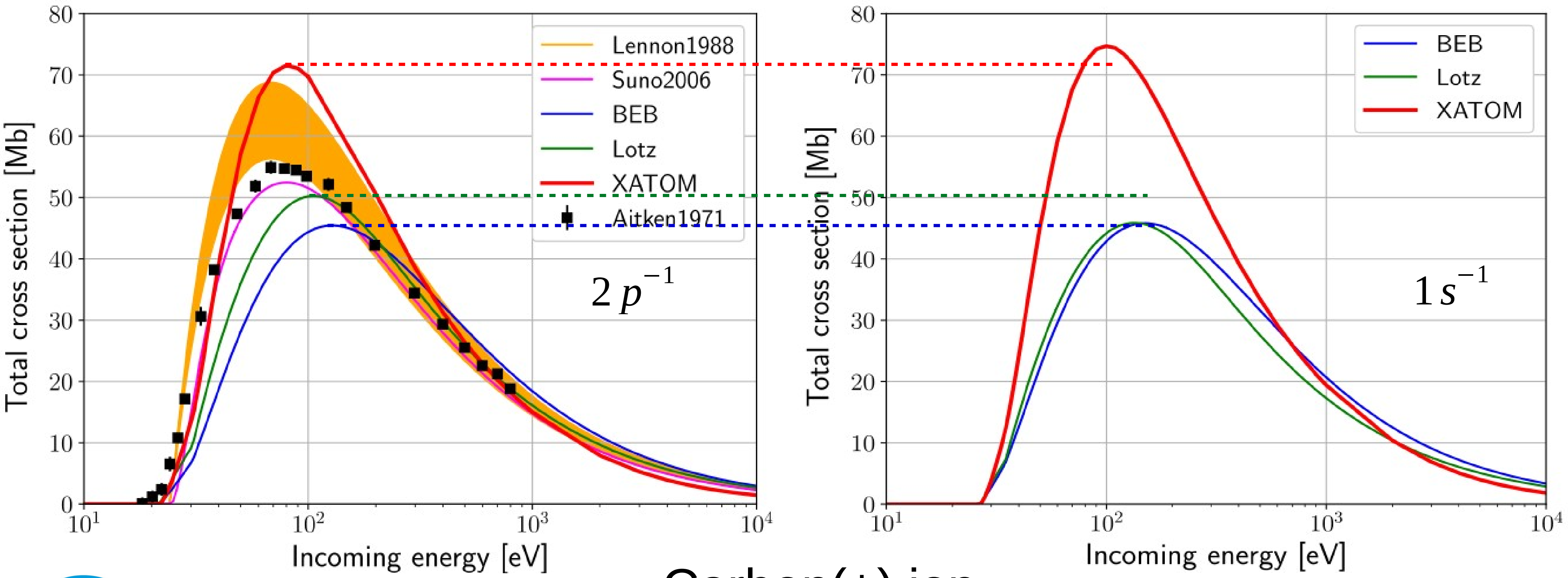
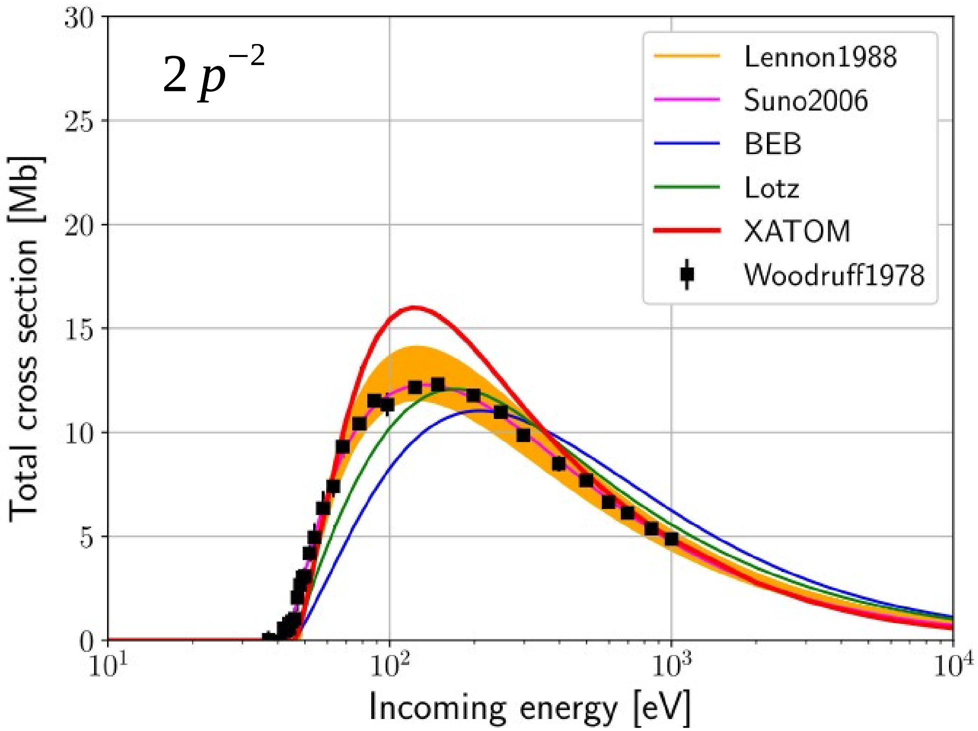


Carbon(+) ion

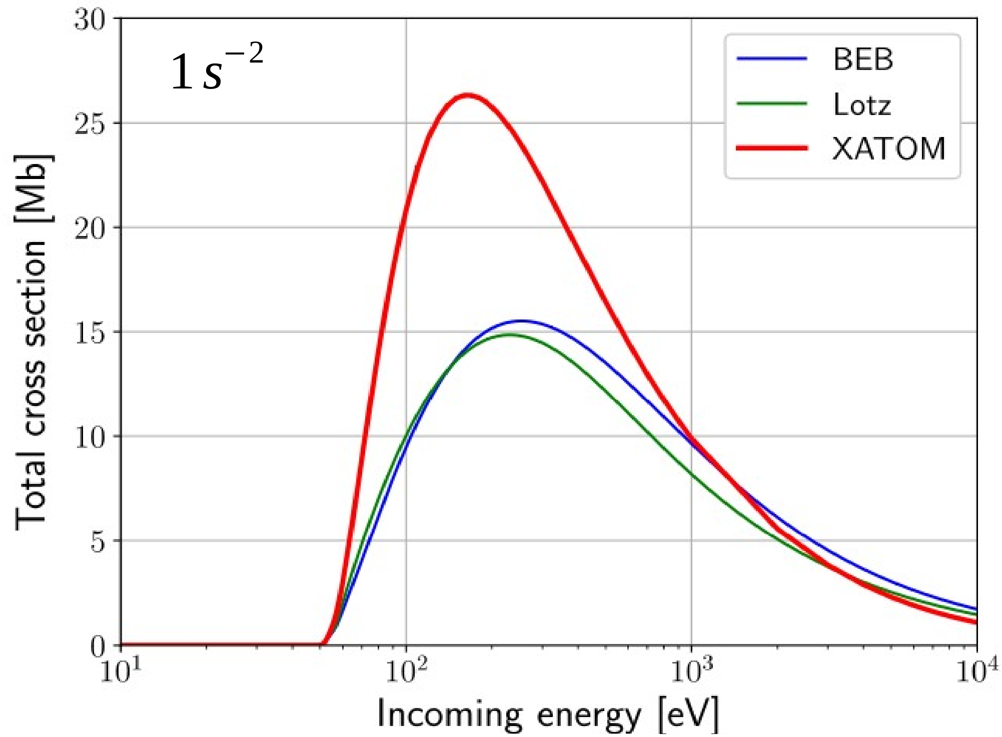
Observations

Reason:

- Largest contributions from valence shells. Core-hole ions have more valence electrons
- Reduced screening in core-hole ions shrinks valence orbitals, which reduces the cross section
- Screening effect decreases for higher incoming energies



Valence holes



Core holes

Carbon(2+) ion

Conclusion & Outlook

- Total cross section results are comparable to other methods
- Exotic electronic configurations in ions can be calculated ab-initio
- Next:
  - Extension to finite temperature
  - Incorporation into irradiated-samples code (XMDYN, nanoplasmas)

Thank you

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HELMHOLTZ
RESEARCH FOR GRAND CHALLENGES

PIER

Universität Hamburg

Total Cross Section

- Comparison made with
  - Experiment [1,2]
  - Lotz formula [3] (semi-empirical)

$$\sigma_{\text{Lotz}} = \sum_{i=1}^{N} a_i q_i \frac{\ln(E/P_i)}{EP_i} \left(1 - b_i \exp\left[-c_i(E/P_i - 1)\right]\right), \quad \text{for } E \geq P_i$$

  - BEB method [4] (currently in use in XMDYN)

$$\sigma_{\text{BEB}} = \frac{S}{t + (u+1)/n} \left[\frac{Q \ln t}{2} \left(1 - \frac{1}{t^2}\right) + (2 - Q)\left(1 - \frac{1}{t} - \frac{\ln t}{t+1}\right) \right]$$

$$t = T/B,\ u = U/B,\ S = 4\pi a_0^2 N (R/B)^2,\ a_0 = 0.52918\ \text{Å},\ R = 13.6057\ \text{eV}$$

[1] M.A. Lennon et al., J. Phys. Chem. Ref. Data **17**, 1285 (1986)
[2] H. Suno and T. Kato, At. Data Nucl. Data Tables, **92** 407 (2006)
[3] W. Lotz, Z. für Phys. **232**, 101-107 (1970)
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Doubly-Differential Cross Section