XATOM: an integrated toolkit for X-ray and atomic physics

Sang-Kil Son[1] and Robin Santra[1,2]

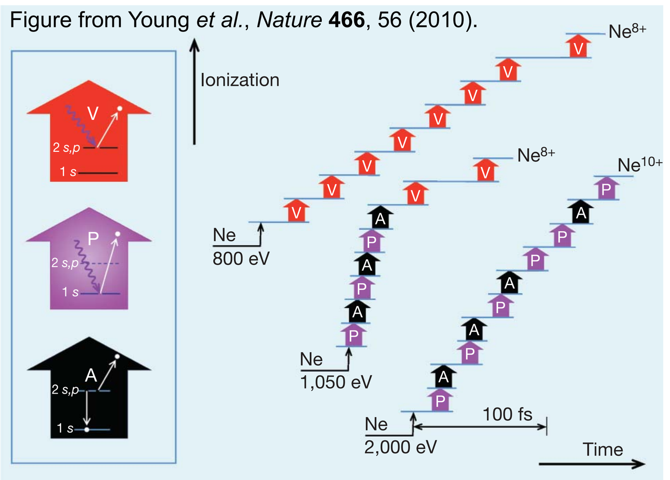
[1]CFEL, DESY, Germany / [2]Department of Physics, University of Hamburg, Germany

Introduction

X-ray free-electron lasers (XFEL) open a new era in science and technology, offering many unique opportunities that have not been conceivable with conventional light sources. Because XFELs produce ultrashort pulses with a very high X-ray photon fluence, materials interacting with XFEL pulses undergo significant radiation damage and possibly become highly ionized. To understand the underlying physics, it is crucial to describe detailed ionization and relaxation dynamics in individual atoms during XFEL pulses. Here we present an integrated toolkit to investigate X-ray-induced atomic processes and to simulate electronic damage dynamics. This XATOM toolkit can handle all possible electronic configurations of all atom/ion species, and calculate physical observables during/after intense X-ray pulses. By use of XATOM, we can explore many exciting XFEL-related phenomena from multiphoton multiple ionization to molecular imaging and warm dense matter formation.

Diagrams of multiphoton absorption mechanisms in Ne induced by ultraintense X-ray pulses

Figure from Young *et al.*, *Nature* **466**, 56 (2010).



Theoretical and numerical details

Hamiltonian and perturbation theory

To treat X-ray–atom interactions, we employ a consistent *ab initio* framework based on nonrelativistic quantum electrodynamics and perturbation theory. For implementation, we use the Hartree-Fock-Slater model with the Latter tail correction.

$$\hat{H} = \hat{H}_{\mathrm{mol}} + \hat{H}_{\mathrm{EM}} + \hat{H}_{\mathrm{int}}$$

$$\hat{H}_{\mathrm{EM}} = \sum_{\mathbf{k},\lambda} \omega_{\mathbf{k}} \hat{a}^\dagger_{\mathbf{k},\lambda}\hat{a}_{\mathbf{k},\lambda}, \quad \omega_{\mathbf{k}} = |\mathbf{k}|/\alpha$$

$$\hat{H}_{\mathrm{int}} = \alpha \int d^3x\, \hat{\psi}^\dagger(\mathbf{x}) \left[\hat{\mathbf{A}}(\mathbf{x}) \cdot \frac{\boldsymbol{\nabla}}{i}\right] \hat{\psi}(\mathbf{x}) + \frac{\alpha^2}{2} \int d^3x\, \hat{\psi}^\dagger(\mathbf{x}) \hat{A}^2(\mathbf{x}) \hat{\psi}(\mathbf{x})$$

$|I\rangle$: initial state, $|F\rangle$: final state

$$\Gamma_{FI} = 2\pi\delta(E_F - E_I) \left| \langle F|\hat{H}_{\mathrm{int}}|I\rangle + \sum_M \frac{\langle F|\hat{H}_{\mathrm{int}}|M\rangle\langle M|\hat{H}_{\mathrm{int}}|I\rangle}{E_I - E_M + i\epsilon} + \cdots \right|^2$$

Rate equation model

To simulate electronic dynamics during intense X-ray pulses, we employ the rate equation approach with all computed cross sections and rates for all possible *n*-hole configurations, and calculate charge state distribution, electron/fluorescence spectra, scattering signals, etc.

$$\frac{d}{dt}P_I(t) = \sum_{I' \neq I}^{\text{all config.}} \left[\Gamma_{I' \to I} P_{I'}(t) - \Gamma_{I \to I'} P_I(t)\right]$$

P Photoabsorption $|I\rangle = |\Psi_0^{N_{\mathrm{el}}}\rangle|N_{\mathrm{EM}}\rangle,\ |F\rangle = |\Psi_F^{N_{\mathrm{el}}}\rangle|N_{\mathrm{EM}}-1\rangle$

$$\sigma_{\mathrm{P}}(i,\omega) = \frac{4}{3}\alpha\pi^2\omega N_i \sum_{l_j=|l_i-1|}^{l_i+1} \frac{l_>}{2l_i+1} \left| \int_0^\infty P_{n_i l_i}(r) P_{\varepsilon l_j}(r)\, r\, dr \right|^2$$

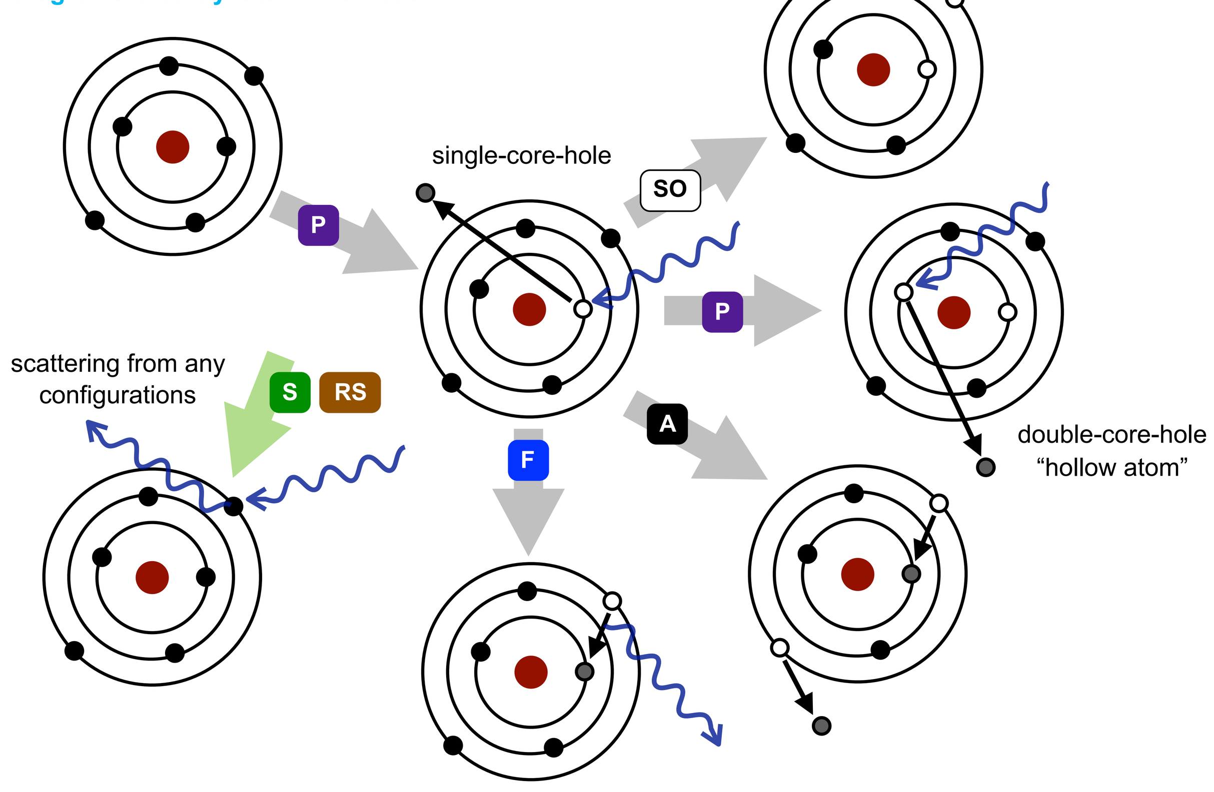
F Fluorescence $|I\rangle = \hat{c}_i|\Phi_0^{N_{\mathrm{el}}}\rangle|0\rangle,\ |F\rangle = \hat{c}_{i'}|\Phi_0^{N_{\mathrm{el}}}\rangle\hat{a}^\dagger_{\mathbf{k}_F,\lambda_F}|0\rangle$

$$\Gamma_{\mathrm{F}}(i,j) = \frac{4}{3}\alpha^3(I_i - I_j)^3 \frac{N_i^{\mathrm{H}} N_j}{4l_j+2} \cdot \frac{l_>}{2l_i+1} \left| \int_0^\infty P_{n_i l_i}(r) P_{n_j l_j}(r)\, r\, dr \right|^2$$

A Auger / Coster–Kronig decay $|I\rangle = \hat{c}_i|\Phi_0^{N_{\mathrm{el}}}\rangle,\ |F\rangle = \hat{c}_a^\dagger\hat{c}_j\hat{c}_{j'}|\Phi_0^{N_{\mathrm{el}}}\rangle$

$$\Gamma_{\mathrm{A}}(i,jj') = \pi\frac{N_i^{\mathrm{H}} N_{jj'}}{2l_i+1} \sum_{L=|l_j-l_{j'}|}^{l_j+l_{j'}} \sum_{S=0}^{1} \sum_{l_{i'}} (2L+1)(2S+1)|M_{LS}(j,j',i,i')|^2$$

Diagrams of X-ray–atom interaction



SO Shake-off process: sudden approximation

$$p_{\mathrm{SO}}(i;I,I') = 1 - \left| \int_0^\infty P_{n_i l_i}(r;I) P_{n_i l_i}(r;I')\, dr \right|^2$$

S Elastic X-ray scattering $|I\rangle = |\Psi_0^{N_{\mathrm{el}}}\rangle|N_{\mathrm{EM}}\rangle$, $|F\rangle = |\Psi_0^{N_{\mathrm{el}}}\rangle\hat{a}^\dagger_{\mathbf{k}_F,\lambda_F}|N_{\mathrm{EM}}-1\rangle$

$$f^0(\mathbf{Q}) = \int \rho(\mathbf{r})\, e^{i\mathbf{Q}\cdot\mathbf{r}}\, d^3r$$

$$\frac{d\sigma_{\mathrm{S}}}{d\Omega}(t) = \sum_I^{\text{all config.}} P_I(t) \left.\frac{d\sigma_{\mathrm{S}}}{d\Omega}\right|_I = \left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{T}} \sum_I^{\text{all config.}} P_I(t)|f_I^0(\mathbf{Q})|^2$$

RS Resonant elastic X-ray scattering (dispersion correction)

$$f(\mathbf{Q},\omega) = f^0(\mathbf{Q}) + f'(\omega) + if''(\omega)$$

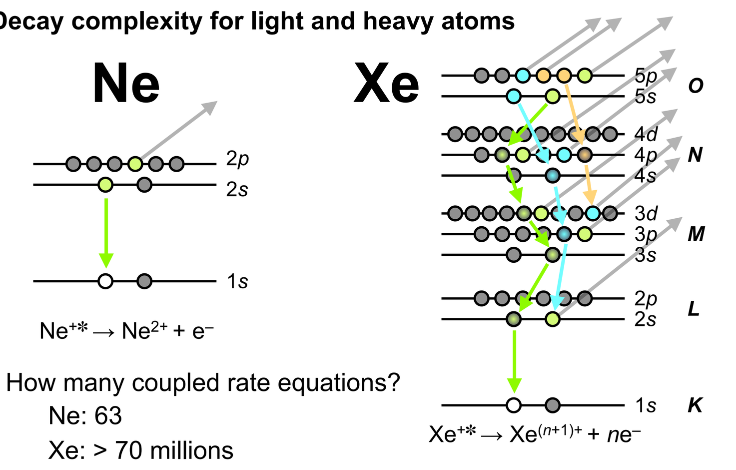
$$f'(\omega) = -\frac{1}{2\pi^2\alpha}\mathcal{P}\int_0^\infty \frac{\omega'^2}{\omega'^2 - \omega^2}\sigma_{\mathrm{P}}(\omega')d\omega'$$

$$f''(\omega) = -\frac{\omega}{4\pi\alpha}\sigma_{\mathrm{P}}(\omega)$$

atom

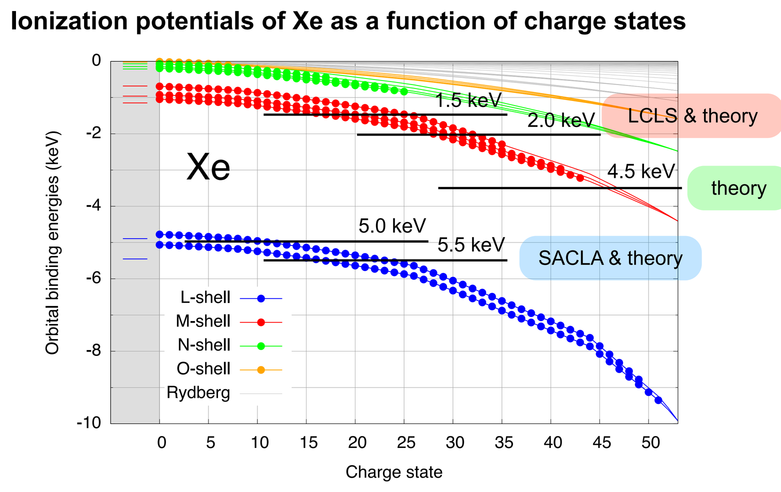
fundamental interaction between atoms and XFEL pulses

Decay complexity for light and heavy atoms



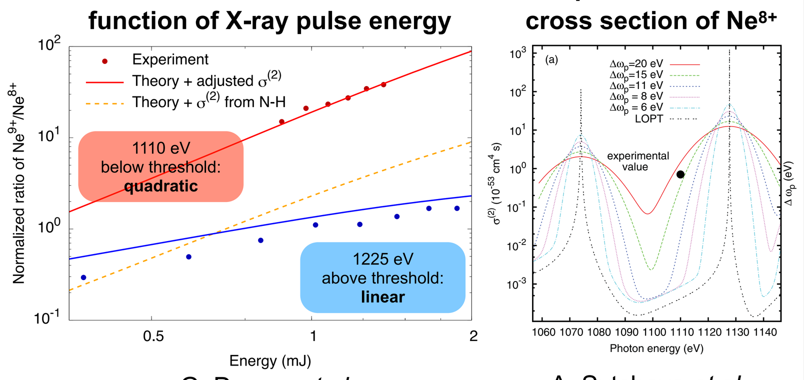
How many coupled rate equations?
Ne: 63
Xe: > 70 millions
Xe + relativity: > 15 billions
Xe + relativity + excitation: infinity

Ionization potentials of Xe as a function of charge states



Ne at LCLS 1110 eV & 1225 eV

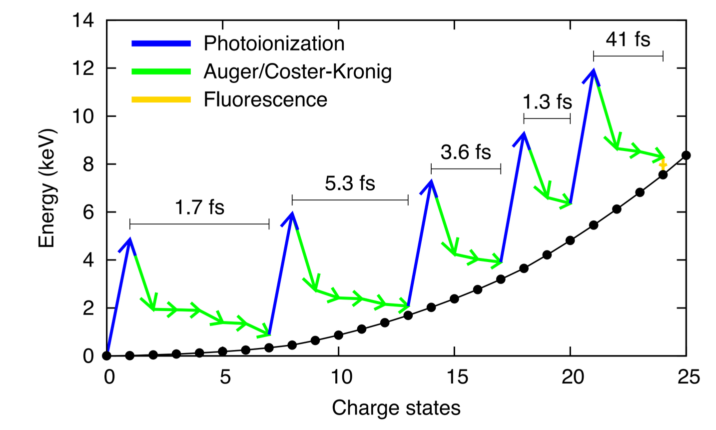
The Ne9+/Ne8+ ratio as a function of X-ray pulse energy | **Two-photon ionization cross section of Ne8+**



G. Doumy *et al.*, *PRL* **106**, 083002 (2011). A. Sytcheva *et al.*, *PRA* **85**, 023414 (2012).

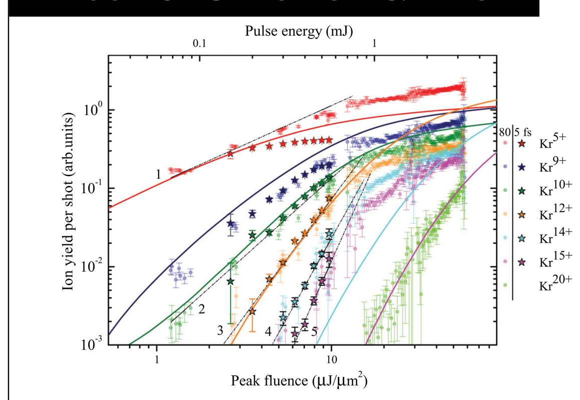
Xe at SACLA 5.5 keV

Deep inner-shell multiphoton multiple ionization mechanism



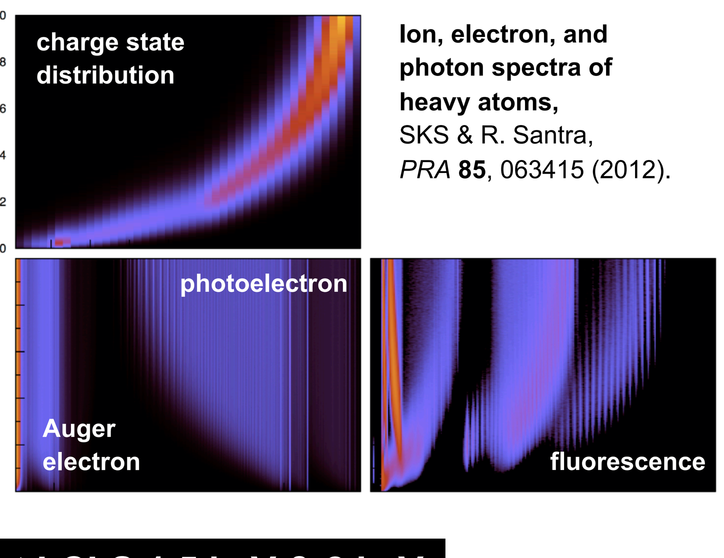
H. Fukuzawa, SKS *et al.*, *PRL* **110**, 173005 (2013).

Kr at LCLS 1.5 keV & 2 keV



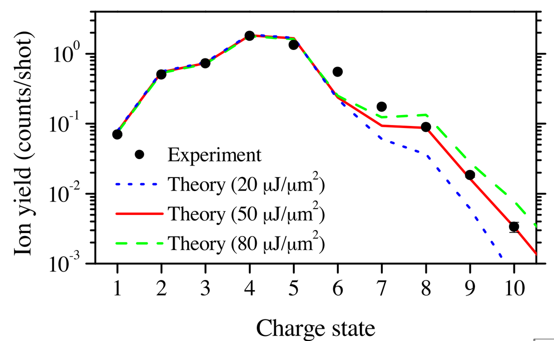
Kr ion yield as a function of X-ray peak fluence, demonstrating REXMI processes, B. Rudek *et al.*, *PRA* **87**, 023413 (2013).

Xe at 4.5 keV



Ion, electron, and photon spectra of heavy atoms, SKS & R. Santra, *PRA* **85**, 063415 (2012).

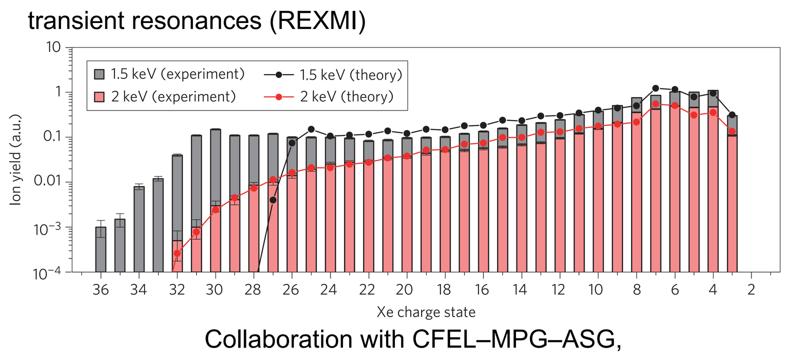
Ar at SACLA 5 keV



Ar charge state distribution, used for XFEL fluence calibration, K. Motomura *et al.*, *JPB* **46**, 164024 (2013).

Xe at LCLS 1.5 keV & 2 keV

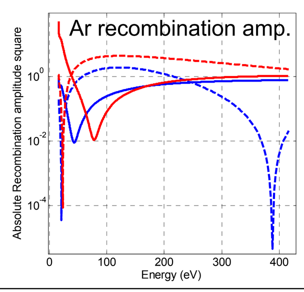
Comparison of experimental and theoretical charge state distributions, revealing new ionization channels via excitation of transient resonances (REXMI)



Collaboration with CFEL–MPG–ASG,
B. Rudek, SKS *et al.*, *Nature Photon.* **6**, 858 (2012).

HHG of rare gases

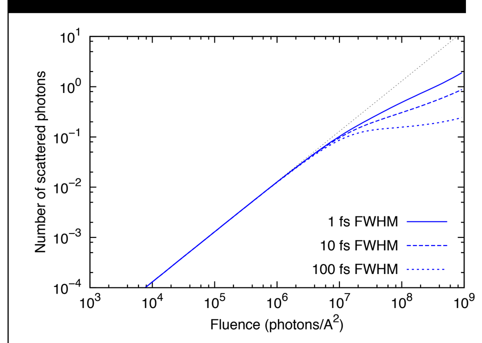
XATOM has been used for calculations of the three-step model of high harmonic generation; Collaboration with CFEL Ultrafast Optics and X-Rays Group, S. Bhardwaj, SKS *et al.*, *PRA* (in press).



molecule

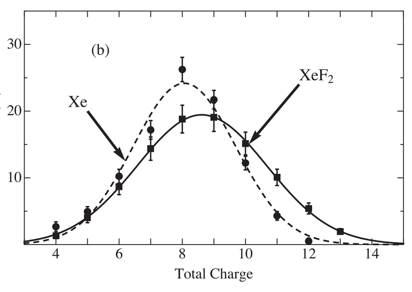
imaging from small molecules to biological macromolecules

Hollow-atom formation



Number of scattered photons for several pulse durations, showing more scatterings for a shorter pulse due to intensity-induced X-ray transparency or frustrated absorption, SKS, L. Young & R. Santra, *PRA* **83**, 033402 (2011).

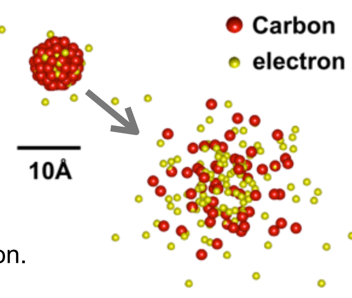
XeF_2 at synchrotron



Comparison of charge state distributions between isolated Xe atom and XeF_2 molecule, R. W. Dunford *et al.*, *PRA* **86**, 033401 (2012).

C_{60} at LCLS

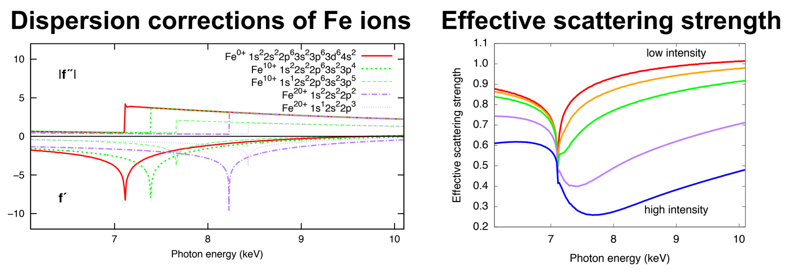
Atomic data calculated by XATOM have been used for molecular dynamics of C_{60}; Collaboration with Z. Jurek, B. F. Murphy *et al.*, in preparation.



MAD at high x-ray intensity

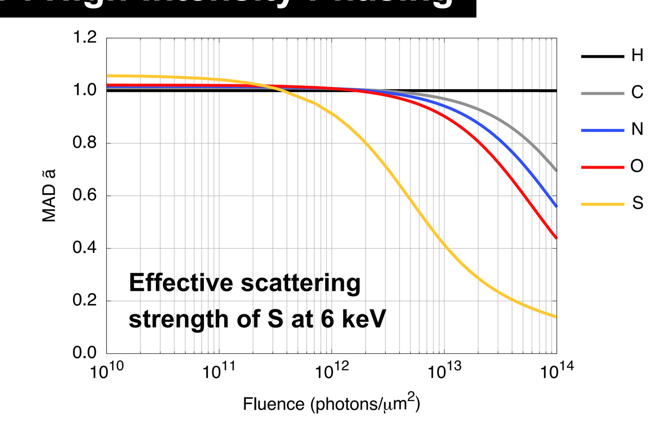
Phasing is crucial to determine the protein structure. Our method has been applied to femtosecond nanocrystallography with XFEL.

Dispersion corrections of Fe ions | **Effective scattering strength**



Basic theory and generalized Karle–Hendrickson equation: SKS, H. Chapman & R. Santra, *PRL* **107**, 218102 (2011).
Configurational and temporal fluctuations and MAD coefficients: SKS, H. Chapman & R. Santra, *JPB* **46**, 164015 (2013).

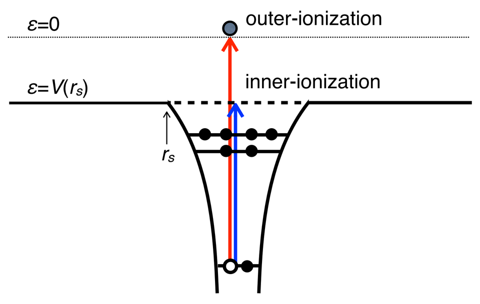
HIP: High-Intensity Phasing



Collaboration with CFEL Coherent Imaging Group, SKS, L. Galli *et al.*, in preparation.

solid

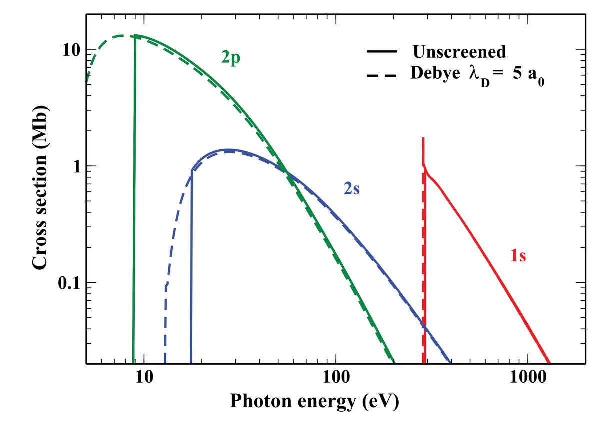
electronic structure in extreme conditions



Atomic picture in dense matter: Muffin-tin-like potential with the Wigner-Seitz radius to describe an environment of an atom within a solid or a plasma

Screening effect

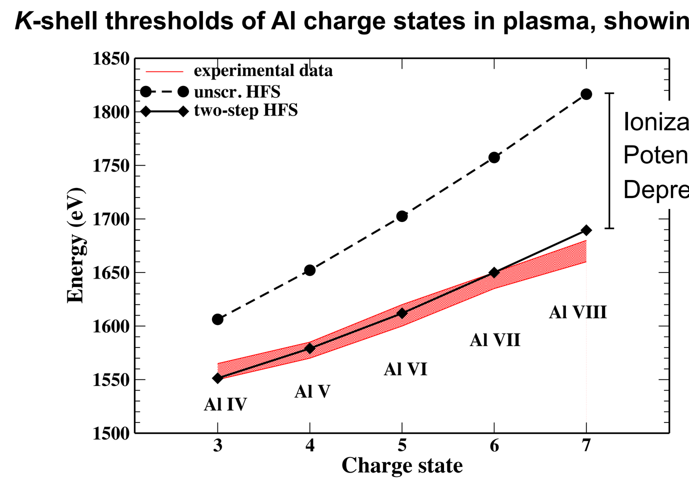
Photoabsorption cross sections for C calculated with the unscreened and Debye-screened HFS models



Model: R. Thiele, SKS *et al.*, *PRA* **86**, 033411 (2012).
Photoelectron spectroscopy: B. Ziaja *et al.*, *JPB* **46**, 164009 (2013).
Magnetic scattering of Co/Pt at FLASH: L. Müller *et al.*, *PRL* **110**, 234801 (2013).

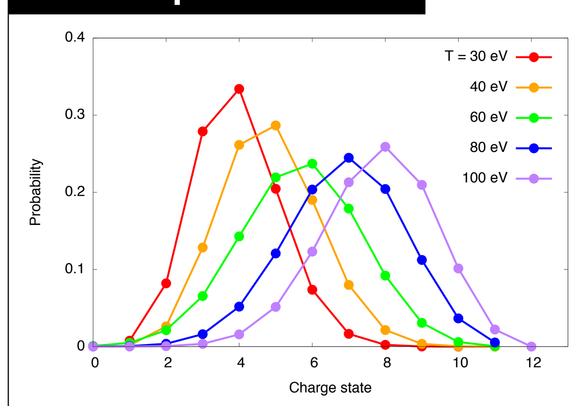
Al plasma at LCLS

***K*-shell thresholds of Al charge states in plasma, showing IPDs**



SKS, R. Thiele *et al.*, submitted.

Two-step HFS model



Probabilities of individual charge states of Al plasma calculated by thermal HFS, selecting a specific configuration and providing a plasma electron density

SKS, R. Thiele *et al.*, in preparation.

Center for Free-Electron Laser Science

CFEL is a scientific cooperation of the three organizations:
DESY – Max Planck Society – University of Hamburg