Effect of screening by external charges on the atomic orbitals and photoinduced processes within the Hartree-Fock-Slater approach

R. Thiele[1] S.-K. Son[1] B. Ziaja-Motyka[1,2] R. Santra[1,3]

[1]CFEL Theory Division, DESY CFEL, Notkestraße 85, 22607 Hamburg, Germany
[2]Institute of Nuclear Physics, Polish Academy of Sciences, Radzikowskiego 152, 31-342 Kraków, Poland
[3]Department of Physics, University of Hamburg, 20355 Hamburg, Germany

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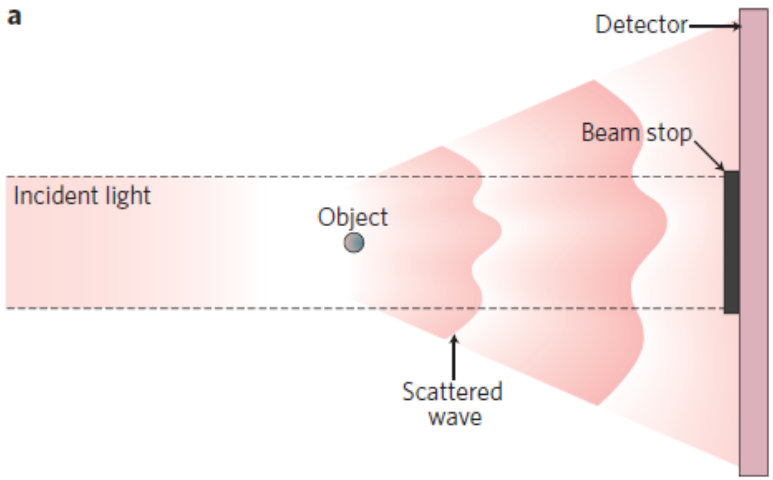
Motivation: Coherent diffraction imaging (CDI) with XFELs

- (X)FELs provide brilliant radiation with extremely high peak brightness in the soft or hard x-ray regime (FLASH, LCLS, etc.)
- One of the unique opportunities offered by XFELs is single-shot imaging of individual macromolecules
- CDI [1] can determine the structure of non-crystallizing biomolecules or other nanoparticles at atomic resolution
- The intense XFEL pulses ionize the molecules $\rightsquigarrow$ plasma screening effects
- Effect of screening on orbital energies, Auger and fluorescence rates, cross sections, etc.

- We use a modified Hartree-Fock Slater approach, implemented in the computational toolkit XATOM [2]
- Effect of charged environment on the photoinduced processes in the context of CDI experiments



[1] H. N. Chapman and K. A. Nugent, Nature Photon. 4, 833 (2010)
[2] S.-K. Son, L. Young, and R. Santra, Phys. Rev. A 83, 033402 (2011)

Effective single-electron Schrödinger equation

$$\left[-\frac{1}{2}\nabla^2 + V_{\text{eff}}(\mathbf{r})\right]\psi(\mathbf{r}) = \varepsilon\psi(\mathbf{r})$$

Effective potential in unscreened HFS approach

$$V_{\text{eff}}^{HFS}(\mathbf{r}) = -\frac{Z}{r} + \int \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\,\mathrm{d}^3r' + V_x(\mathbf{r})$$

Electron density

$$\rho(\mathbf{r}) = \sum_i^{N_{elec}} \psi_i^\dagger(\mathbf{r})\psi_i(\mathbf{r})$$

Slater exchange potential

$$V_x(\mathbf{r}) = -\frac{3}{2}\left[\frac{3}{\pi}\rho(\mathbf{r})\right]^{1/3}$$

[1] J. C. Slater, Phys. Rev. 81, 385 (1951)
[2] S.-K. Son, L. Young, and R. Santra, Phys. Rev. A 83, 033402 (2011)

Effective potential in generalized Debye screened HFS approach

$$V_{\text{eff}}^{xD}(\mathbf{r}) = -\frac{Z e^{-r/\lambda_D}}{r} + \int \frac{[\rho_b(\mathbf{r}') + \bar{n}_e - \bar{Z}_i \bar{n}_i g_{ii}(r')] e^{-|\mathbf{r}-\mathbf{r}'|/\lambda_D}}{|\mathbf{r}-\mathbf{r}'|} \, \mathrm{d}^3 r' + V_x^D(\mathbf{r})$$

Debye-screened exchange potential [1,2]

$$V_x^D(\mathbf{r}) = V_x(\mathbf{r}) F(\alpha)$$

Correction factor by Robinson [1]

$$F(\alpha) = 1 - \frac{\alpha^2}{6} - \frac{4}{3}\alpha \tan^{-1}\left(\frac{2}{\alpha}\right) + \frac{\alpha^2}{2}\left(1 + \frac{\alpha^2}{12}\right) \ln\left|1 + \frac{4}{\alpha^2}\right|$$

where $\alpha = 1/(\lambda_D k_F) = \kappa_D / k_F$, with the Fermi momentum k_F

[1] J. E. Robinson, F. Bassani, R. S. Knox, and J. R. Schrieffer, Phys. Rev. Lett. 9, 215 (1962)
[2] L. M. Sachs, Acta Crystallographica 22, 931 (1967)

$\overline{n}_e$ and $\overline{n}_i$: number densities of external electrons and ions
$\overline{Z}_i$: effective charge of ions

Radial distribution function $g_{ii}(r)$: fit formula [1,2] for parametrization

$$g_{ii}(r) = 1 + a \cdot \mathrm{e}^{-b \cdot r} \sin(c \cdot r - d)$$

with fit parameters a, b, c, d in units of Å^{-1}

Please note:

- For $\lambda_D \to \infty$ we get the high temperature limit
- For $\lambda_D \to 0$ ($\lambda_D \approx a_0$) the Debye theory becomes incorrect
- For degenerate systems ($\Theta_e < 1$ or low temperatures), the temperature T_e in λ_D changes to $\sqrt{T_e^2 + T_F^2}$ with the Fermi temperature T_F

[1] A. A. Broyles, J. Chem. Phys. 35, 493 (1961)
[2] A. A. Broyles, J. Chem. Phys. 34, 1068 (1961)

3. Uniform quasineutral plasmas with $g_{ii} = 1$

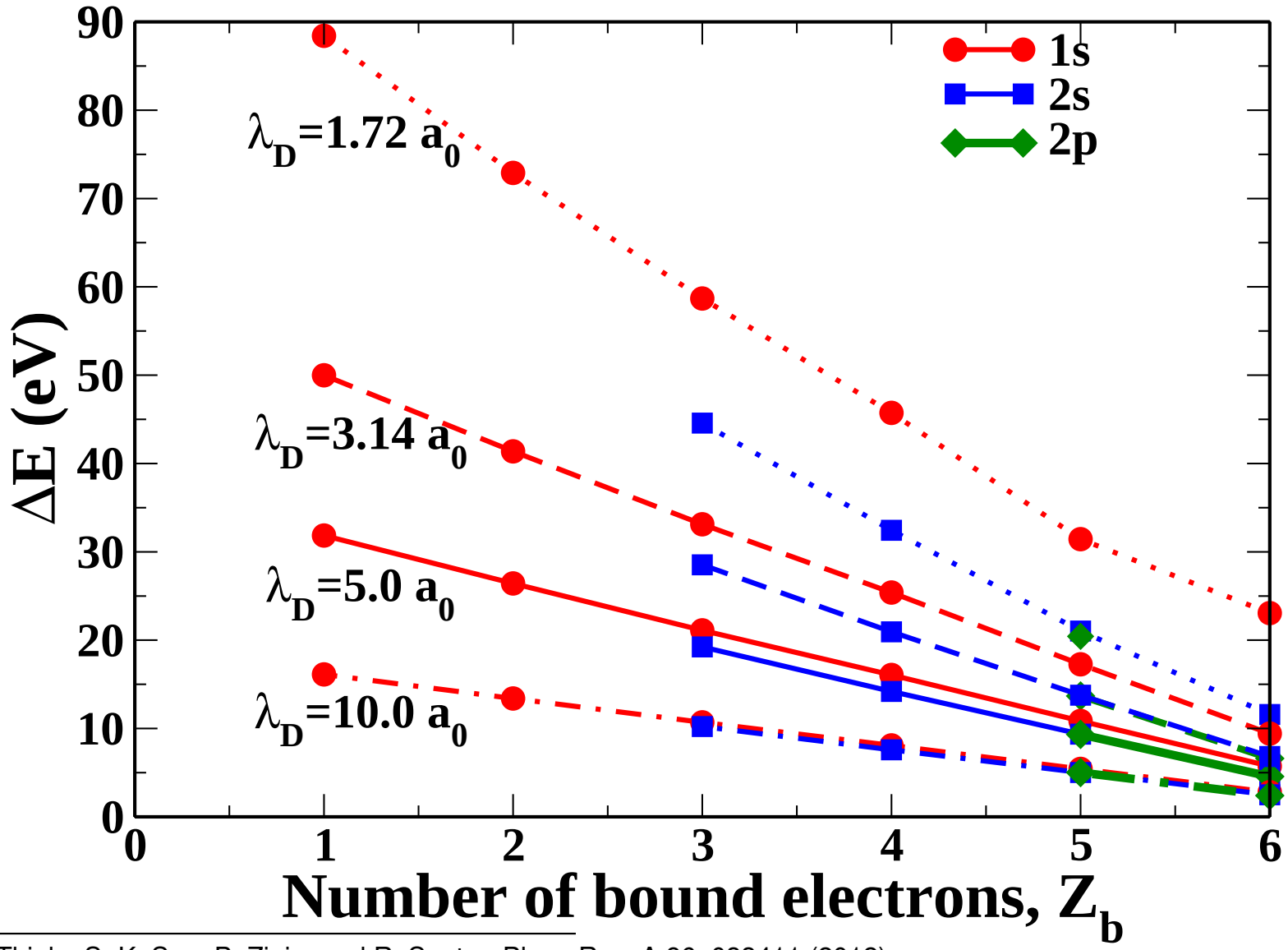
$$V_{\text{eff}}^{D}(\mathbf{r}) = -\frac{Z e^{-r/\lambda_D}}{r} + \int \frac{\rho(\mathbf{r}') e^{-|\mathbf{r}-\mathbf{r}'|/\lambda_D}}{|\mathbf{r}-\mathbf{r}'|} \, \mathrm{d}^3 r' + V_x^D(\mathbf{r})$$

4. High temperature limit of the generalized Debye model

$$V_{\text{eff}}^{IS}(\mathbf{r}) = -\frac{Z}{r} + \int \frac{\rho(\mathbf{r}') + \overline{n}_e - \overline{Z}_i \overline{n}_i g_{ii}(r')}{|\mathbf{r}-\mathbf{r}'|} \, \mathrm{d}^3 r' + V_x(\mathbf{r})$$

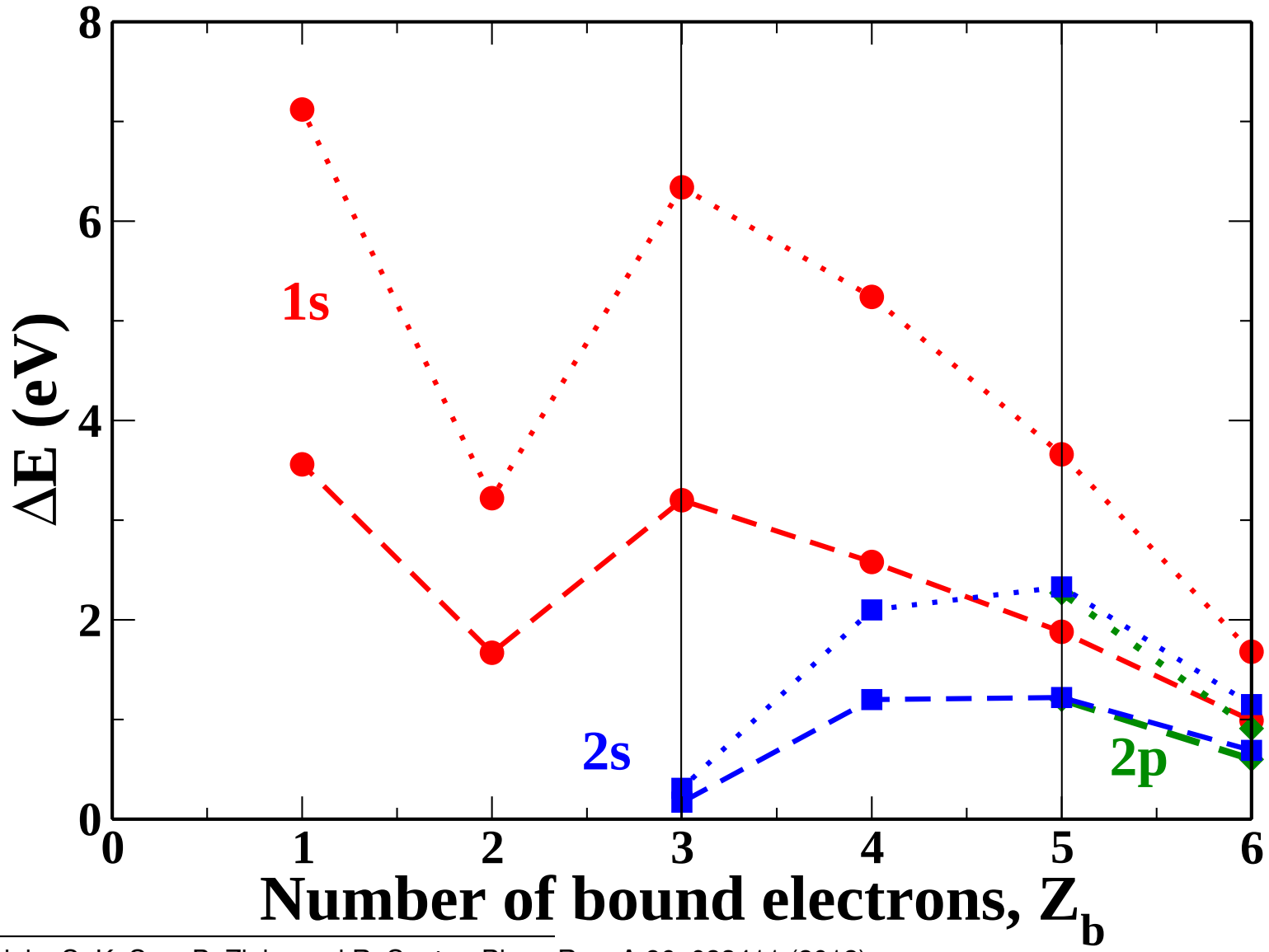
[1] R. Thiele, S.-K. Son, B. Ziaja, and R. Santra, Phys. Rev. A 86, 033411 (2012)

Debye screened HFS method for an uniform plasma: Orbital energy shift for carbon[1]



[1] R. Thiele, S.-K. Son, B. Ziaja, and R. Santra, Phys. Rev. A 86, 033411 (2012)

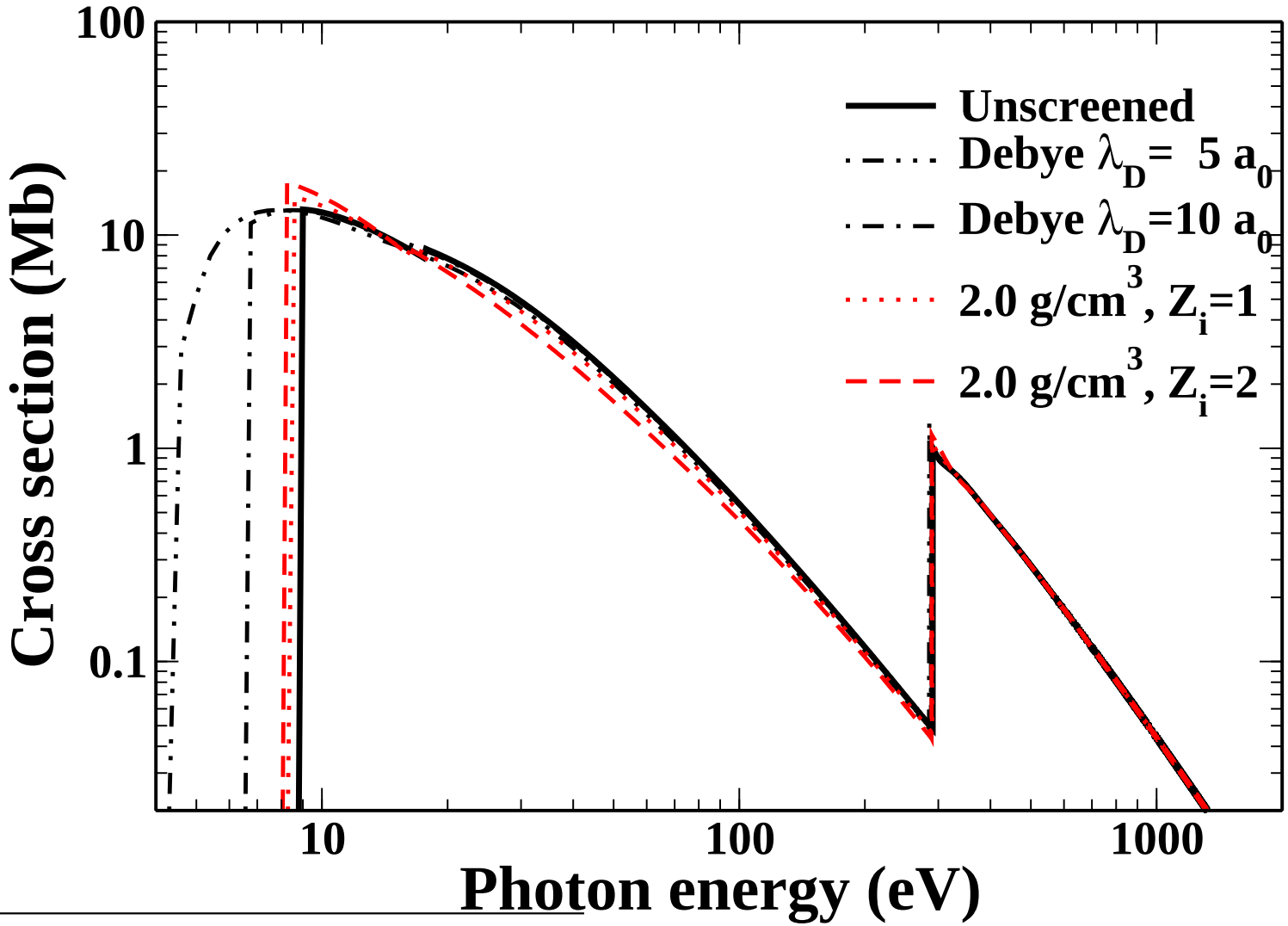
High temperature limit: Orbital energy shift for carbon @ $n_i = 2.0$ g/cm^3 [1]



[1] R. Thiele, S.-K. Son, B. Ziaja, and R. Santra, Phys. Rev. A 86, 033411 (2012)

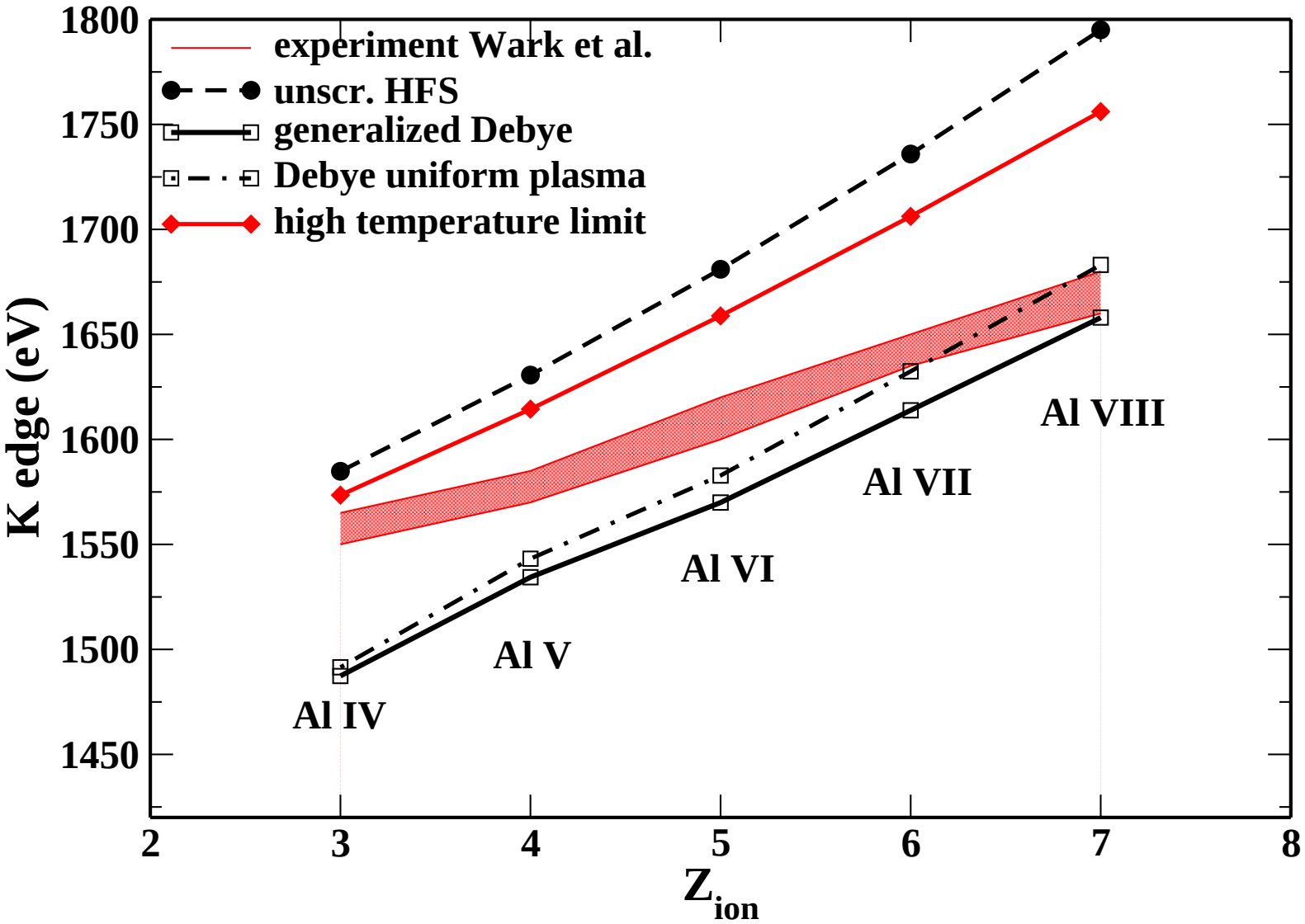
Total photoabsorption cross sections for carbon[1]

Please note: calculations are made for the uniform Debye HFS and respectively the high temperature limit!



[1] R. Thiele, S.-K. Son, B. Ziaja, and R. Santra, Phys. Rev. A 86, 033411 (2012)

K edge for aluminum



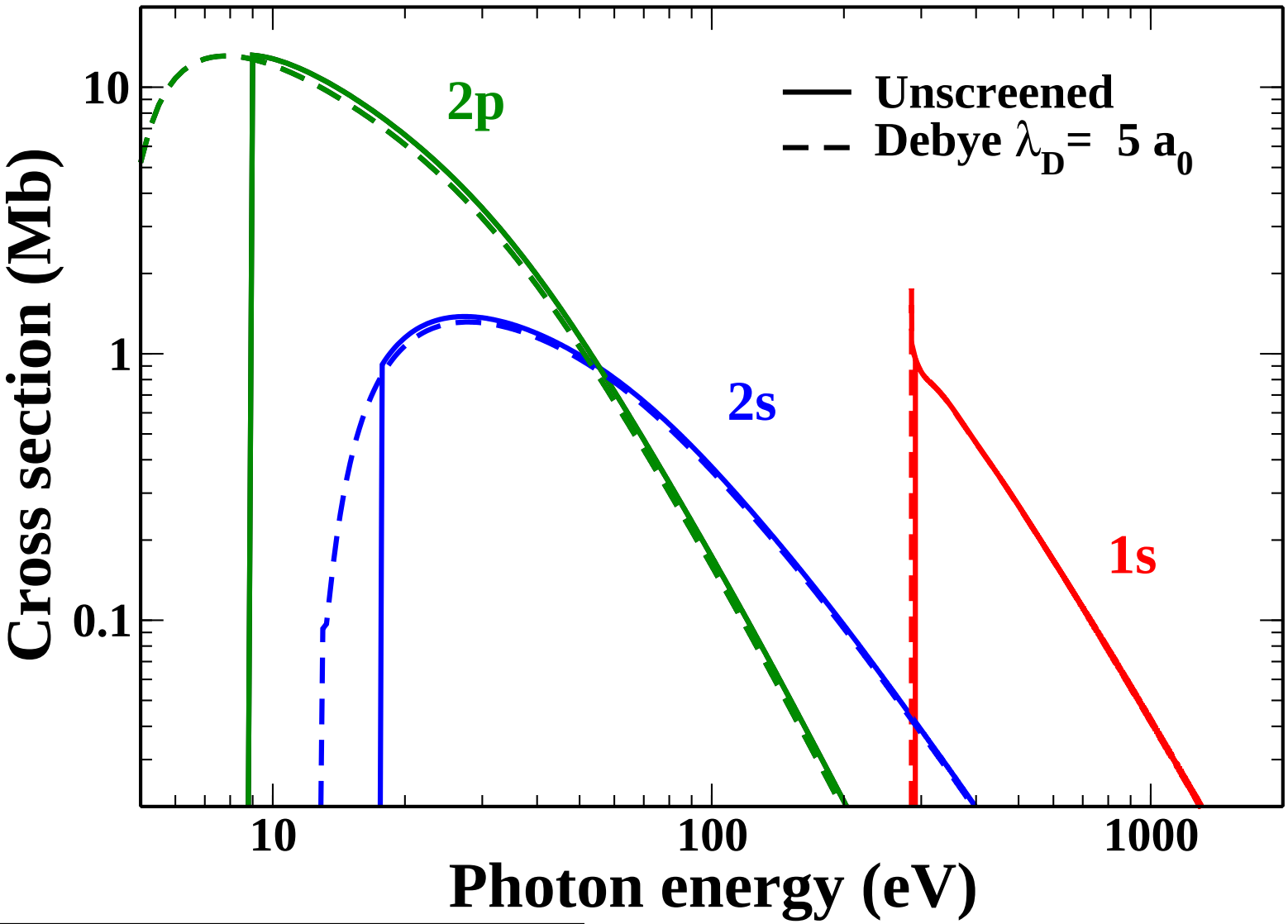
[1] S.M. Vinko *et al.*, Nature 482, 59 (2012)
[2] O. Ciricosta *et al.*, Phys. Rev. Lett. 109, 065002 (2012)

Summary and outlook

- We have extended the standard HFS model and its numerical implementation within the XATOM code to include screening effects, using the generalized Debye model accounting for screening by thermalized free electrons and cold background ions
- In particular, we discussed the limiting cases:
  - **A** of quasi-neutral plasma with uniform ion background, only accounting for screening by thermalized free electrons
  - **B** in the high-temperature limit
- We have calculated orbital energy shifts and photoabsorption cross sections for carbon and 1*s* orbital energies for aluminum embedded in a charged system

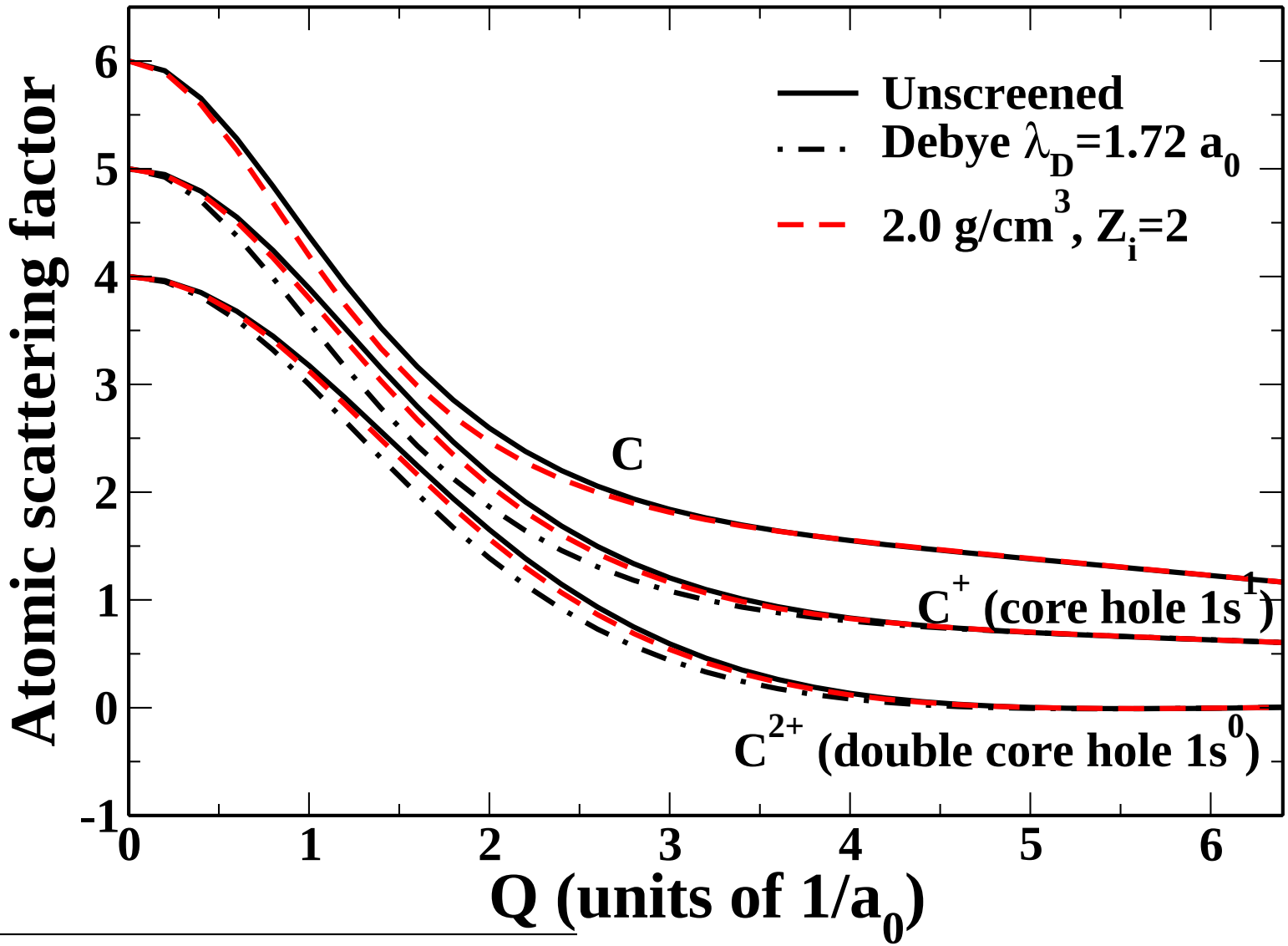
- Development of atomic calculations, yielding distribution of bound and free electrons at a given temperature $\rightsquigarrow$ finite temperature Hartree-Fock approach
- Extend our study to include self-consistent adjustment of screening parameters during the system evolution

Carbon photoabsorption cross sections for each subshell[1]



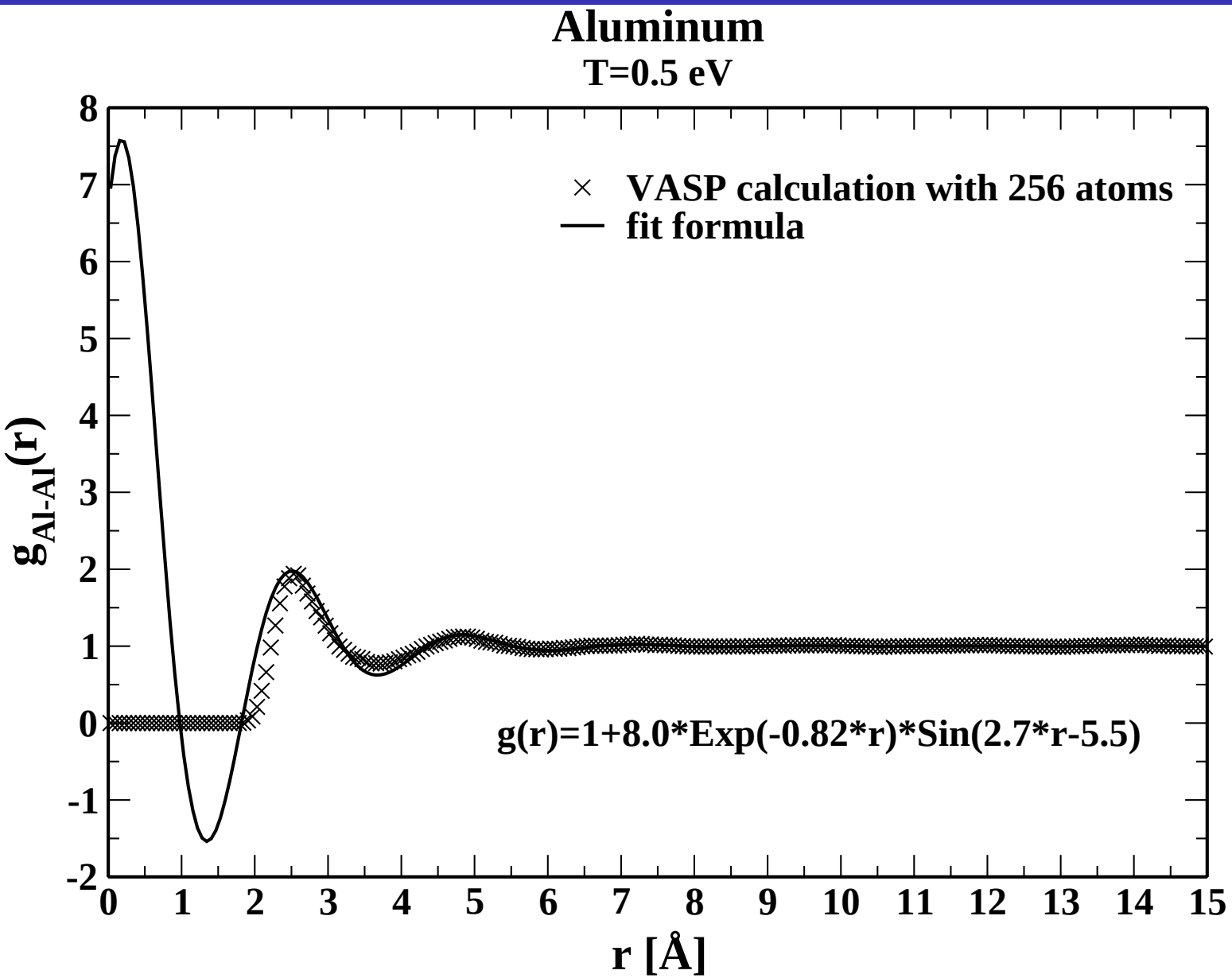
[1] R. Thiele, S.-K. Son, B. Ziaja, and R. Santra, Phys. Rev. A 86, 033411 (2012)

Atomic scattering factor for carbon[1]



[1] R. Thiele, S.-K. Son, B. Ziaja, and R. Santra, Phys. Rev. A 86, 033411 (2012)

Radial distribution function for aluminum plasma



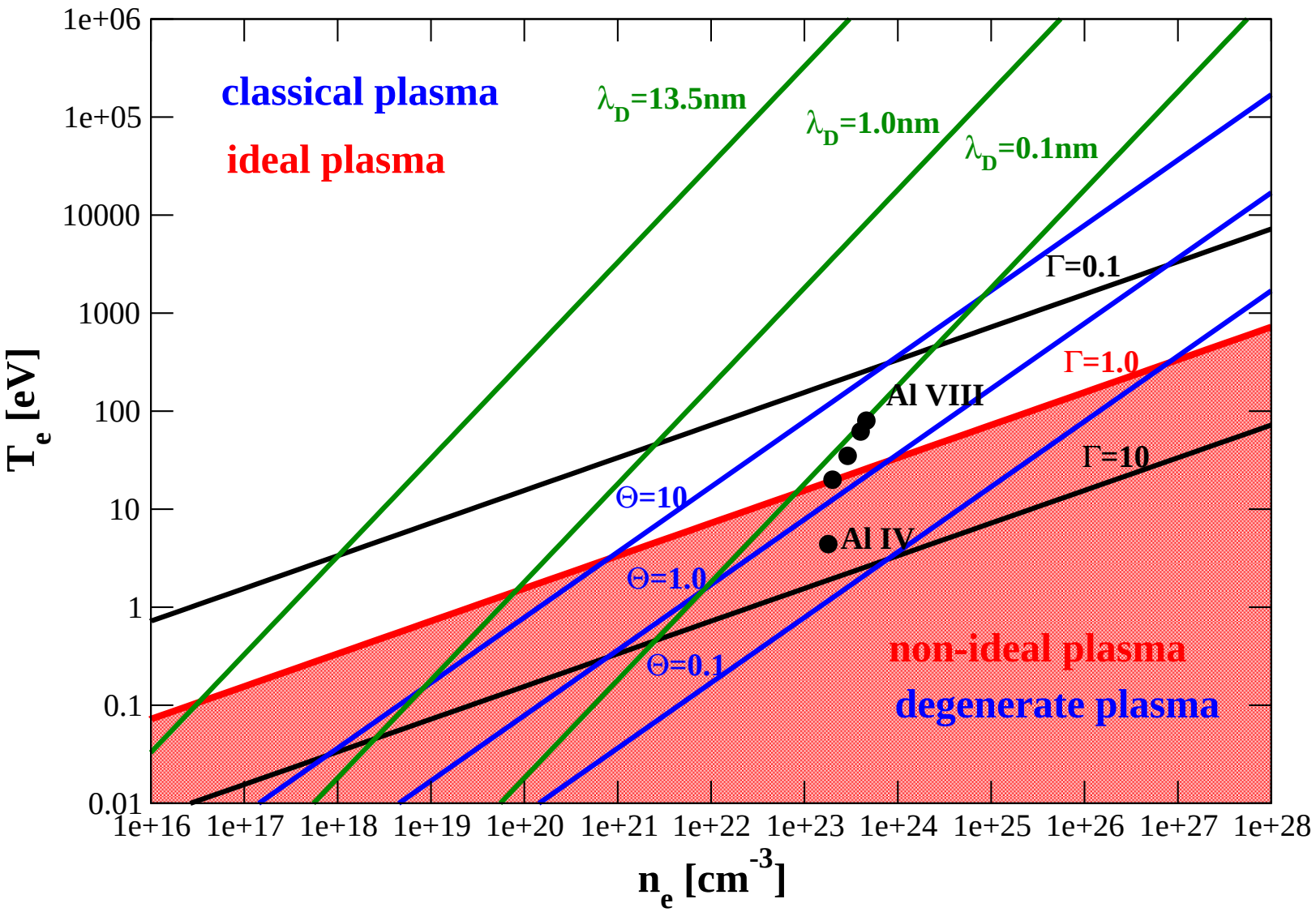
Experimental parameters [1,2]

line	Z_{ion}	n_e [10^{23}cm^{-3}]	T_e [eV]	Θ	Γ	λ_D [Å]	$E_{1s,a}$ [eV]	$E_{1s,exp}$ [eV]
IV	3	1.8	4.4	0.38	2.98	0.37	1610	1550 1565
V	4	2.0	20.0	1.60	0.68	0.74	1655	1570 1585
VI	5	2.9	35.0	2.19	0.44	0.82	1705	1600 1620
VII	6	4.0	62.0	3.13	0.28	0.93	1773	1635 1650
VIII	7	4.6	80.0	3.68	0.22	0.98	1826	1660 1680

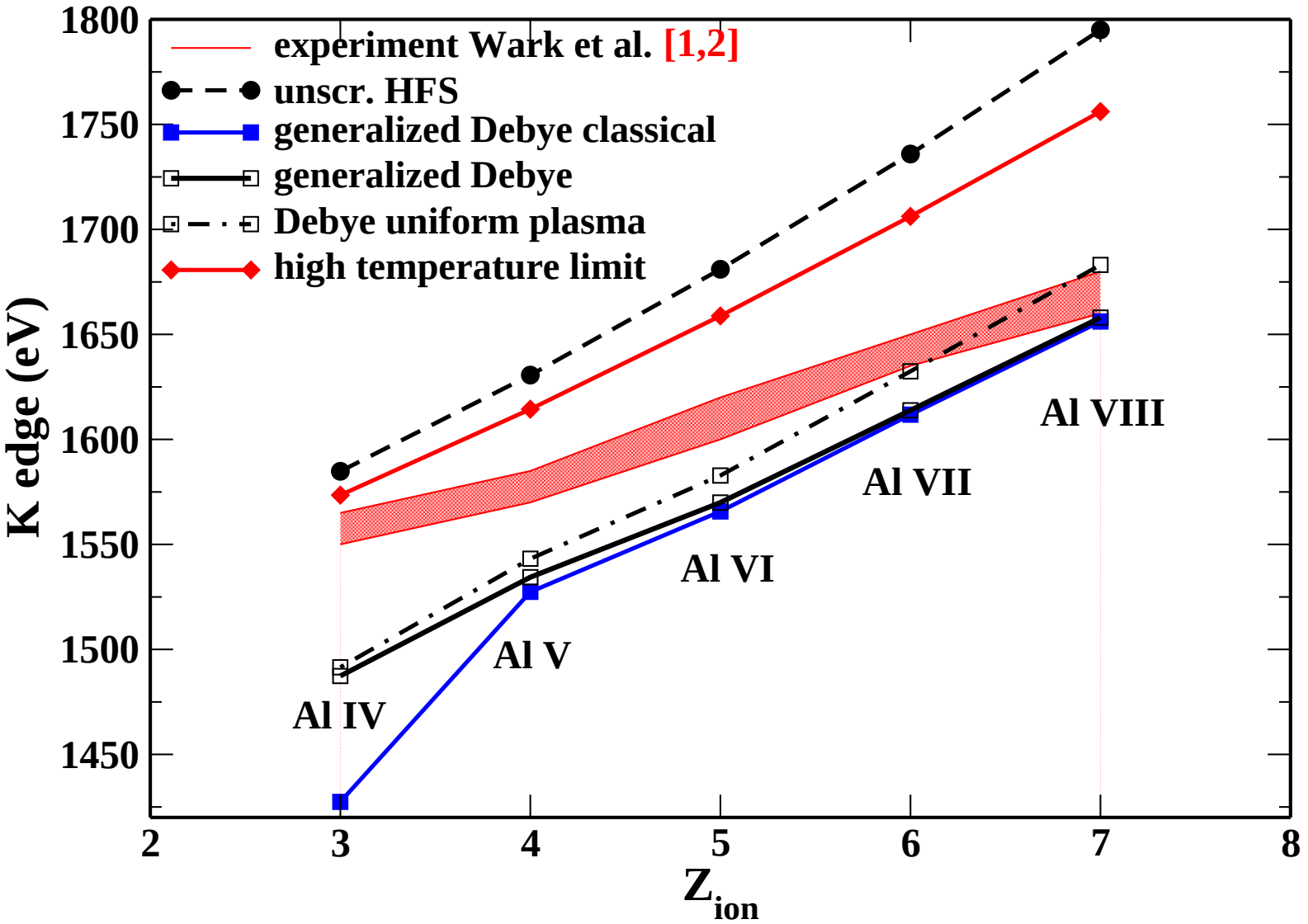
- Ion density is constant: $n_i = 6.02 \times 10^{23}\mathrm{cm}^{-3}$
- $E_{1s,a}$ [eV] are the theoretical energies for K-edge given by J. Wark
- $E_{1s,exp}$ [eV] are the experimental ranges for K-edge given by J. Wark
- Al IV is: $1s^2 2s^2 2p^6 3s^0 3p^0$

[1] S.M. Vinko *et al.*, Nature 482, 59 (2012)
[2] O. Ciricosta *et al.*, Phys. Rev. Lett. 109, 065002 (2012)

Density-temperature plane



K edge for aluminum with PRELIMINARY experimental data



[1] S.M. Vinko *et al.*, Nature 482, 59 (2012)
[2] O. Ciricosta *et al.*, Phys. Rev. Lett. 109, 065002 (2012)