Quantum-mechanical calculation of ionization potential lowering in dense plasmas

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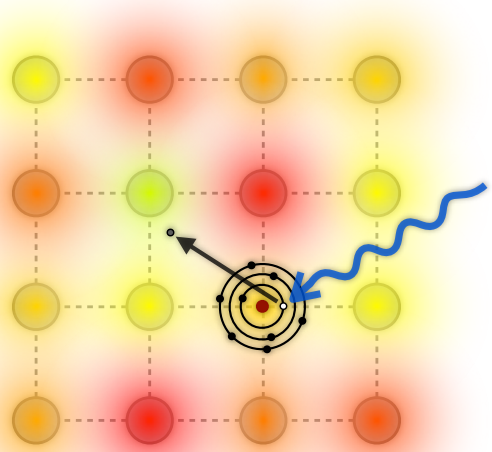
Radiative Properties of Hot Dense Matter (RPHDM)
Vienna, Austria / September 29–October 3, 2014



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

DESY

Collaboration

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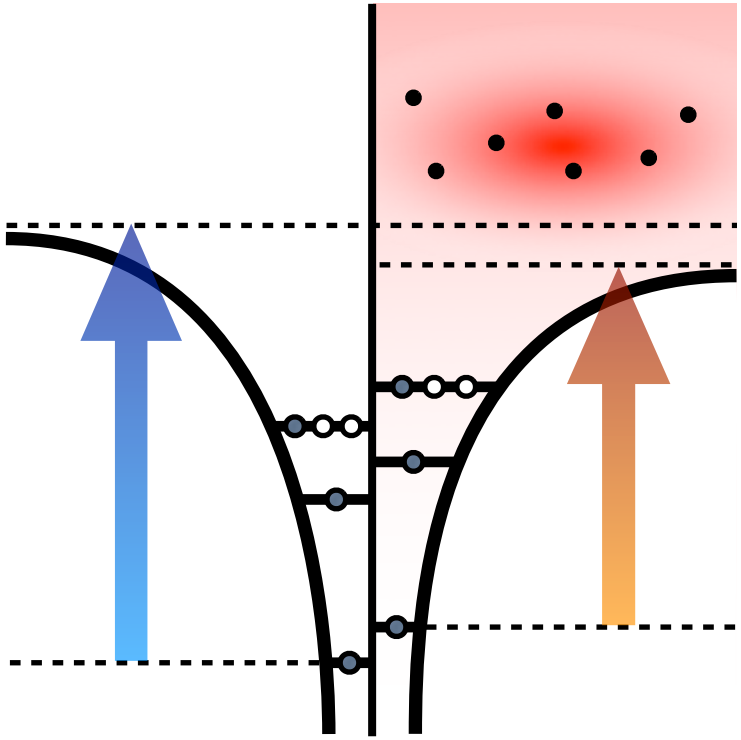
Ionization Potential Depression (IPD)

isolated atom

- Coulomb potential by the nucleus
- screening by bound electrons



solid / plasma

Dense environment

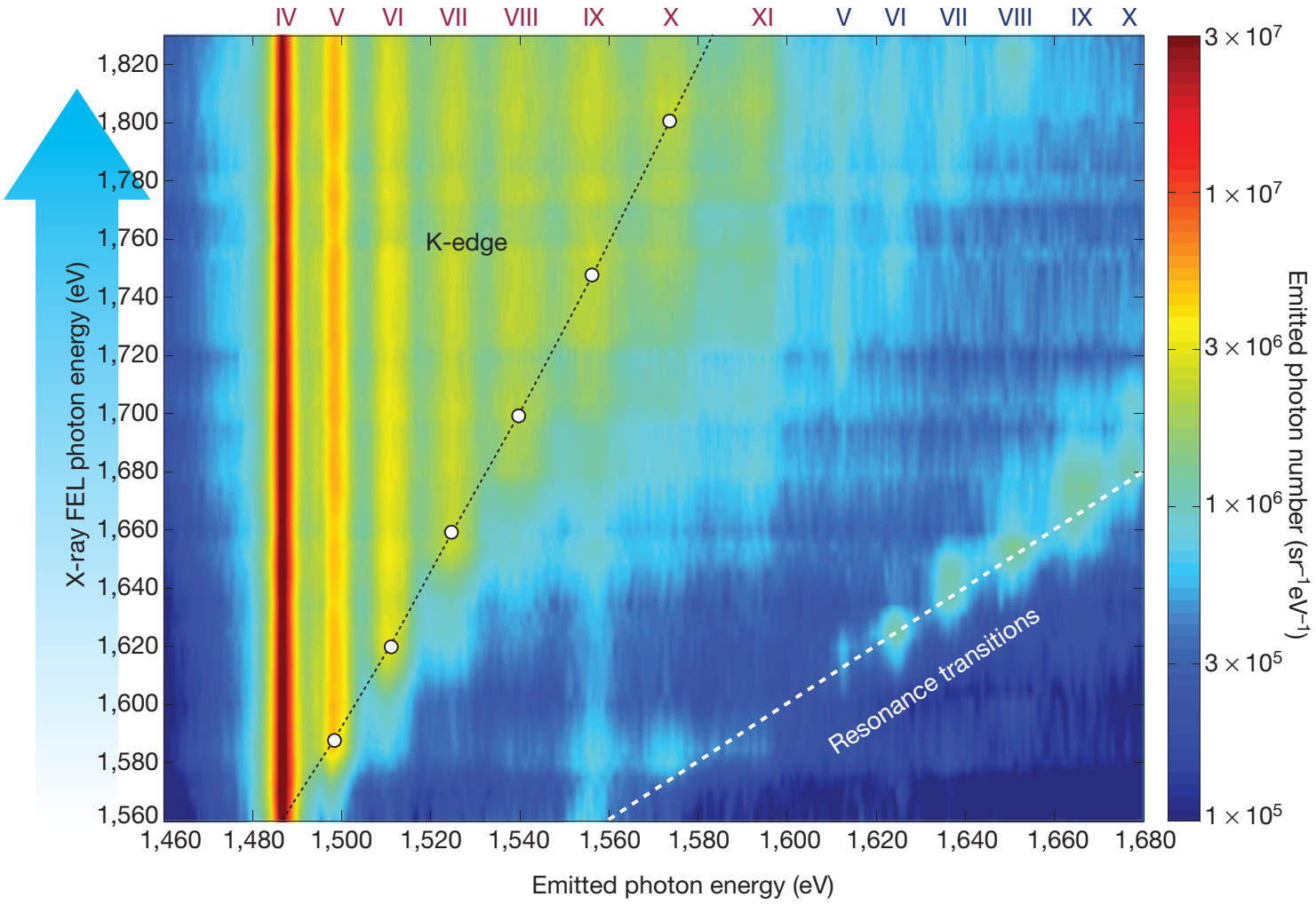
+ screening by free electrons

+ pressure ionization

➔ IP lowering

One of the most fundamental physics for atomic processes in a dense plasma

Warm dense Al plasma (LCLS experiment)

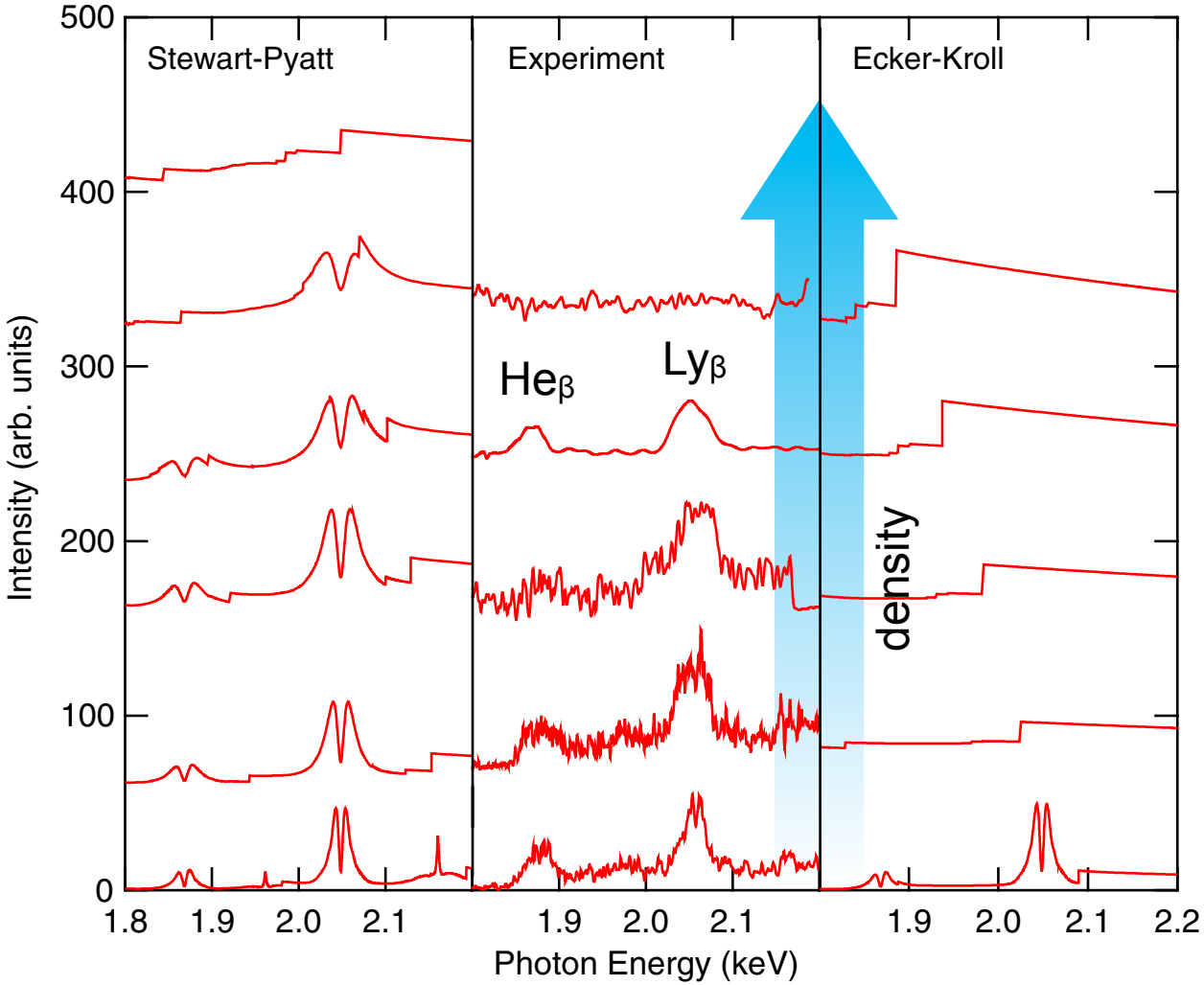


$T < 200$ eV
solid density
(2.7 g/cm^3)

Vinko *et al.*, *Nature* **482**, 59 (2012);

Ciricosta *et al.*, *Phys. Rev. Lett.* **109**, 065002 (2012).

Hot dense Al plasma (Orion experiment)



T ~ 700 eV
0.5~4×solid density

Hoarty *et al.*,
PRL **110**, 265003
(2013).

IPD models

- EK model: Ecker & Kröll, *Phys. Fluids* **6**, 62 (1963)
- SP model: Stewart & Pyatt, *Astrophys. J.* **144**, 1203 (1966)
  - both provide a simple analytic formula for IPD
  - valid only for valence ionization
- LCLS experiment (2012): **modified EK** **SP** 
- Orion experiment (2013): **EK**  **SP**
- Strong need for a rigorous and consistent theoretical approach to IPD

Two-step model: overview

- Two-step model based on
  - Quantum-mechanical calculation: Hartree-Fock-Slater method
  - muffin-tin approximation
  - pseudocontinuum calculation

First step

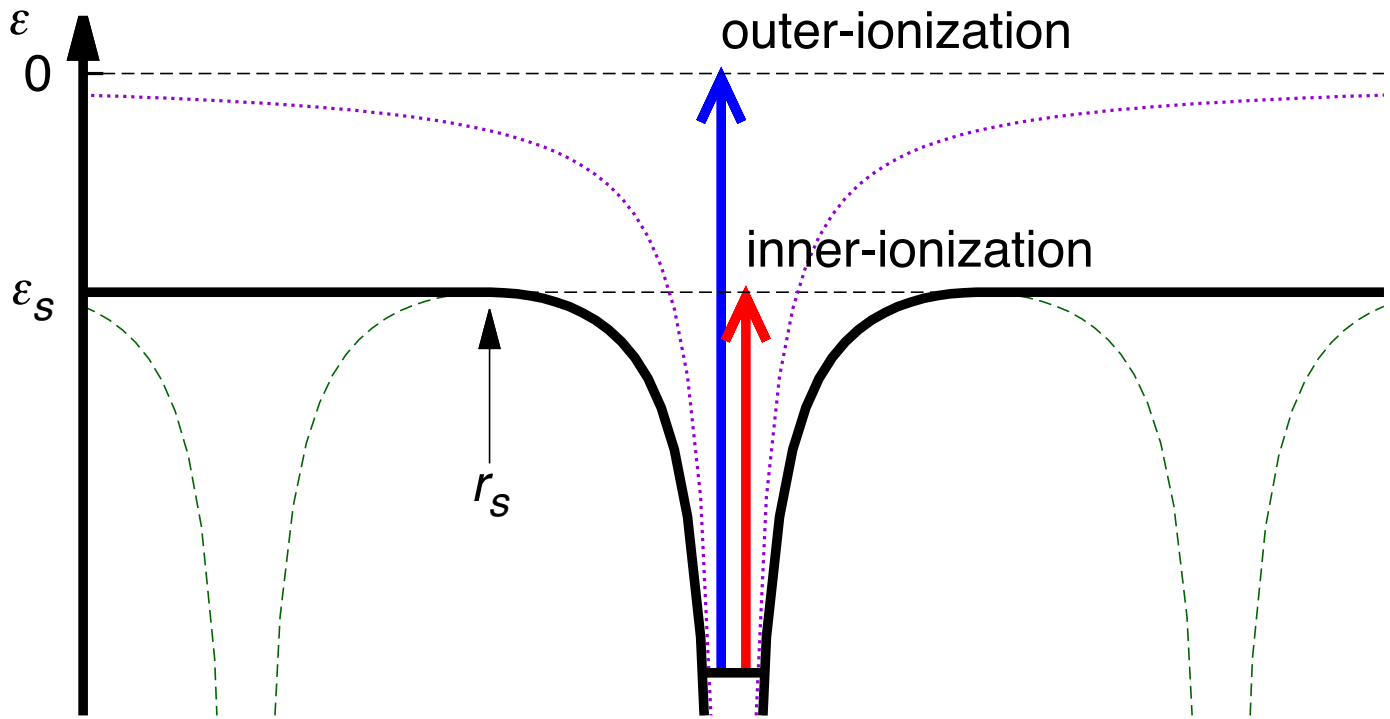
Average-atom calculation

Second step

fixed-configuration calculation

Son, Thiele, Jurek, Ziaja & Santra, *Phys. Rev. X* **4**, 031004 (2014).

Atom in a solid or a plasma



Wigner-Seitz radius: $r_s = \left(\frac{3}{4\pi n_i} \right)^{1/3}$

Inner-ionization potential:

$$E_j = \varepsilon_s - \varepsilon_j$$

Hartree-Fock-Slater method

- Solve the Schrödinger equation (SE)

$$\hat{H} = -\frac{1}{2}\nabla^2 + V(\mathbf{r})$$

$$\hat{H}\psi(\mathbf{r}) = \varepsilon\psi(\mathbf{r})$$

- HFS potential inside the WS radius / muffin-tin flat potential outside

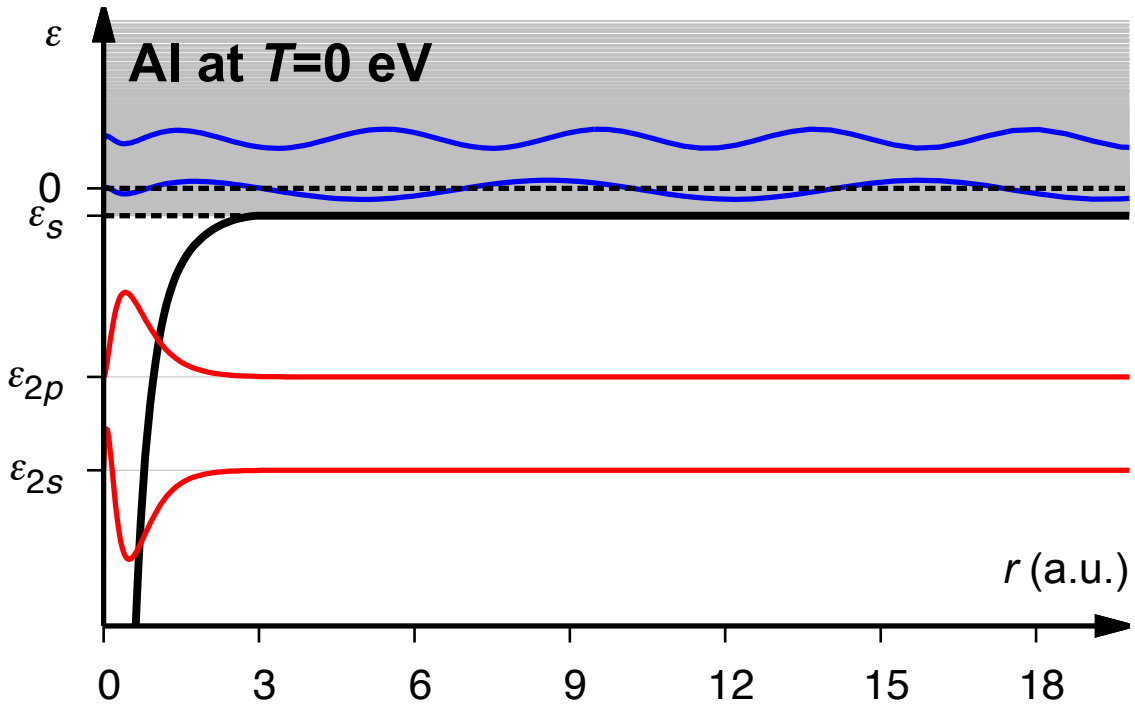
$$V(\mathbf{r}) = \begin{cases} -\dfrac{Z}{r} + \displaystyle\int_{r' \le r_s} d^3r' \, \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + V_{\mathrm{x}}[\rho(\mathbf{r})] & \text{for } r \le r_s, \\ V(r_s) & \text{for } r > r_s, \end{cases}$$

- Slater exchange potential

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

Continuum-state calculation

Radial SE: $$\left[-\frac{1}{2}\frac{d^2}{dr^2}+\frac{l(l+1)}{2r^2}+V(r)\right]u_{nl}(r)=\varepsilon_{nl}u_{nl}(r)$$



- non-uniform radial grids: N_r = 200, 0 ≤ r ≤ 100 a.u.
- partial waves: 0 ≤ l ≤ 30
- 6200 radial eigenstates
  - $\varepsilon < \varepsilon_s$: bound
  - $\varepsilon \geq \varepsilon_s$: pseudocontinuum
- no boundary condition at r_s
- widely used in strong-field atomic physics calculations

First step: average-atom

- Grand-canonical ensemble at a finite temperature *T*
- Solve SE with $\hat{H} = -\frac{1}{2}\nabla^2 + V\left[\rho(\mathbf{r}, T)\right]$
- Electronic density (bound & continuum states)

$$\rho(\mathbf{r}, T) = \sum_p \left|\psi_p(\mathbf{r})\right|^2 \bar{n}_p(\mu, T)$$

- Fermi-Dirac distribution $\bar{n}_p(\mu, T) = \frac{1}{e^{(\varepsilon_p - \mu)/T} + 1}$
- Chemical potential

$$Z - \sum_p \left(\int_{r \leq r_s} d^3 r \ \left|\psi_p(\mathbf{r})\right|^2 \right) \bar{n}_p(\mu, T) = 0$$

Average-atom variants

- quantum-mechanical (HFS or LDA) vs. semiclassical (Thomas-Fermi)
- configuration vs. superconfiguration vs. screened hydrogenic model
- muffin-tin approximation vs. extended model
  - Cauble *et al.*, *Phys. Rev. A* **29**, 3280 (1984).
- QM with configuration
  - Liberman, *Phys. Rev. B* **20**, 4981 (1979).
  - Blenski & Ishikawa, *Phys. Rev. E* **51**, 4869 (1995).
  - Sahoo *et al.*, *Phys. Rev. E* **77**, 046402 (2008).
  - Johnson, Nilsen & Cheng, *Phys. Rev. E* **86**, 036410 (2012).
- QM with superconfiguration
  - Pain, Dejonghe & Blenski, *JQSRT* **99**, 451 (2006); *J. Phys. A* **39**, 4659 (2006).

First step: average-atom calculation

- Assumption: thermalized hot electrons; cold ions
- Input parameter: element (Z), temperature (T), and solid density (via r_s)
- Self-consistently determined: orbitals, orbital energies, electron density, muffin-tin flat potential, and chemical potential

Al
solid
density

T	$\bar{Q}$	ε_{1s}	$\cdots$	ε_s	μ
10	+3.01	−1541.14		−11.03	−12.57
30	+3.95	−1579.28		−12.46	−58.67
40	+4.83	−1606.37		−13.19	−85.66
60	+5.67	−1657.70		−14.33	−145.43
80	+6.87	−1702.23		−15.15	−211.69

ensemble-averaged charge and orbital energies

First step: more than average

Q	Configuration	Probability	E_K	$E_{K\alpha}$
+5	$1s^2 2s^1 2p^4 3s^0 3p^1$	0.0193	1618.3	1497.7
	$1s^2 2s^2 2p^3 3s^0 3p^1$	0.0187	1623.1	1500.3
	$1s^2 2s^2 2p^4 3s^0 3p^0$	0.0174	1578.7	1486.7
+6	$1s^2 2s^1 2p^3 3s^0 3p^1$	0.0376	1658.1	1511.6
	$1s^2 2s^1 2p^4 3s^0 3p^0$	0.0349	1618.3	1497.7
	$1s^2 2s^2 2p^3 3s^0 3p^0$	0.0339	1623.1	1500.3
	$1s^2 2s^2 2p^2 3s^0 3p^1$	0.0205	1663.5	1514.5
	$1s^2 2s^1 2p^3 3s^1 3p^0$	0.0139	1656.0	1511.3
+7	$1s^2 2s^1 2p^3 3s^0 3p^0$	0.0681	1666.3	1512.8
	$1s^2 2s^1 2p^2 3s^0 3p^1$	0.0413	1705.4	1527.8
	$1s^2 2s^2 2p^2 3s^0 3p^0$	0.0371	1671.9	1515.8
	$1s^2 2s^0 2p^3 3s^0 3p^1$	0.0189	1699.3	1524.5
	$1s^2 2s^0 2p^4 3s^0 3p^0$	0.0175	1660.9	1509.9
	$1s^2 2s^1 2p^2 3s^1 3p^0$	0.0153	1705.4	1527.9
	$1s^2 2s^2 2p^1 3s^0 3p^1$	0.0120	1711.7	1531.2
+8	$1s^2 2s^1 2p^2 3s^0 3p^0$	0.0747	1718.7	1530.0
	$1s^2 2s^0 2p^3 3s^0 3p^0$	0.0342	1712.3	1526.7
	$1s^2 2s^1 2p^1 3s^0 3p^1$	0.0241	1758.5	1546.5
	$1s^2 2s^2 2p^1 3s^0 3p^0$	0.0217	1725.1	1533.4
	$1s^2 2s^0 2p^2 3s^0 3p^1$	0.0207	1751.6	1542.9
+9	$1s^2 2s^1 2p^1 3s^0 3p^0$	0.0437	1775.1	1549.6
	$1s^2 2s^0 2p^2 3s^0 3p^0$	0.0375	1768.0	1545.9
	$1s^2 2s^0 2p^1 3s^0 3p^1$	0.0121	1808.2	1564.1
+10	$1s^2 2s^0 2p^1 3s^0 3p^0$	0.0219	1827.4	1568.1
	$1s^2 2s^1 2p^0 3s^0 3p^0$	0.0106	1835.2	1572.1

Al at T=80 eV

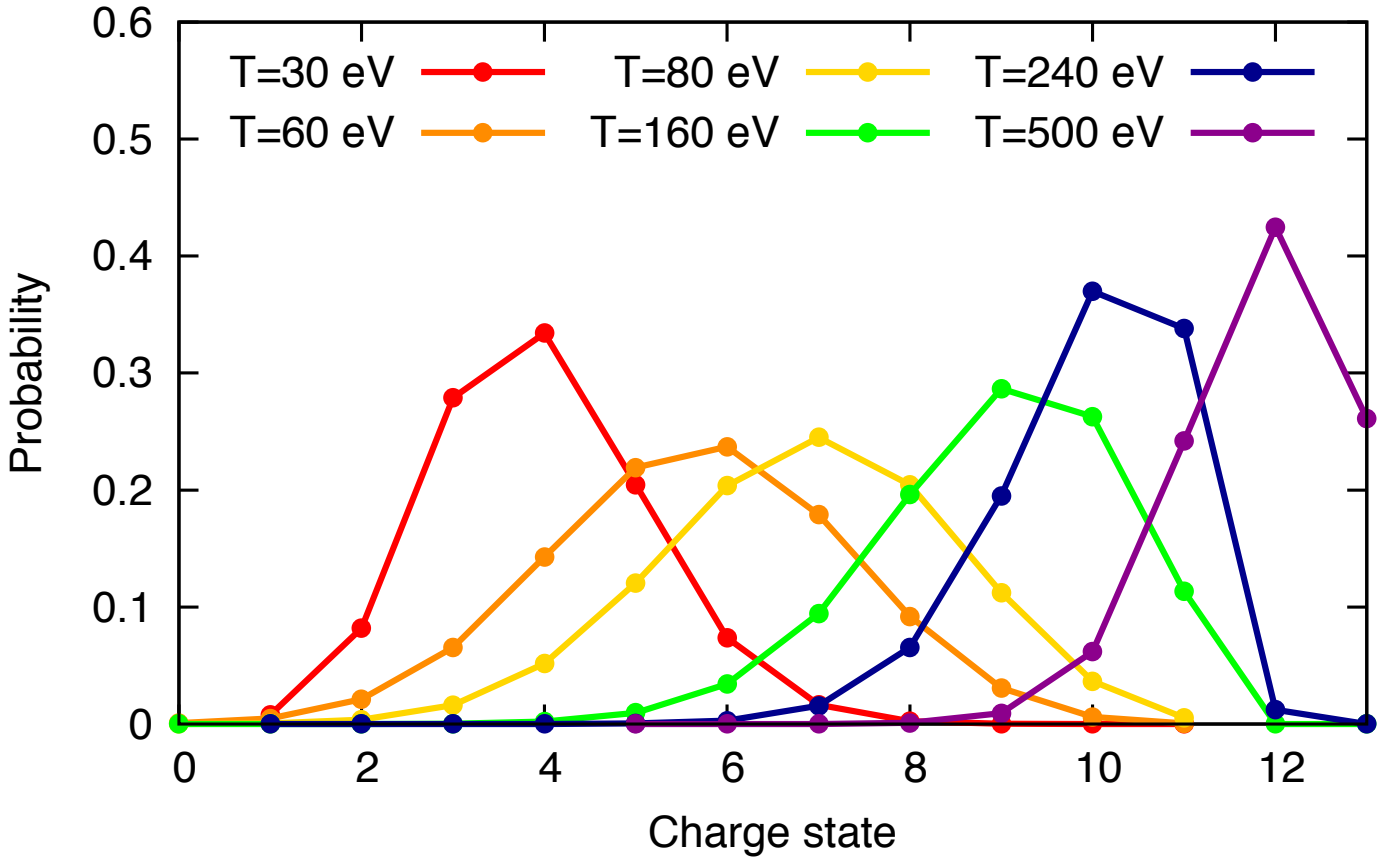
From the grand-canonical ensemble, probability distributions calculated for given bound-state configurations

$$P_{[n_b]} = \sum_{\{n_p\}=\{n_b;n_{p'}\}} P_{\{n_p\}}$$

$$= \prod_{b}^{\text{bound}} \frac{e^{-(\varepsilon_b-\mu)n_b/T}}{1+e^{-(\varepsilon_b-\mu)/T}}$$

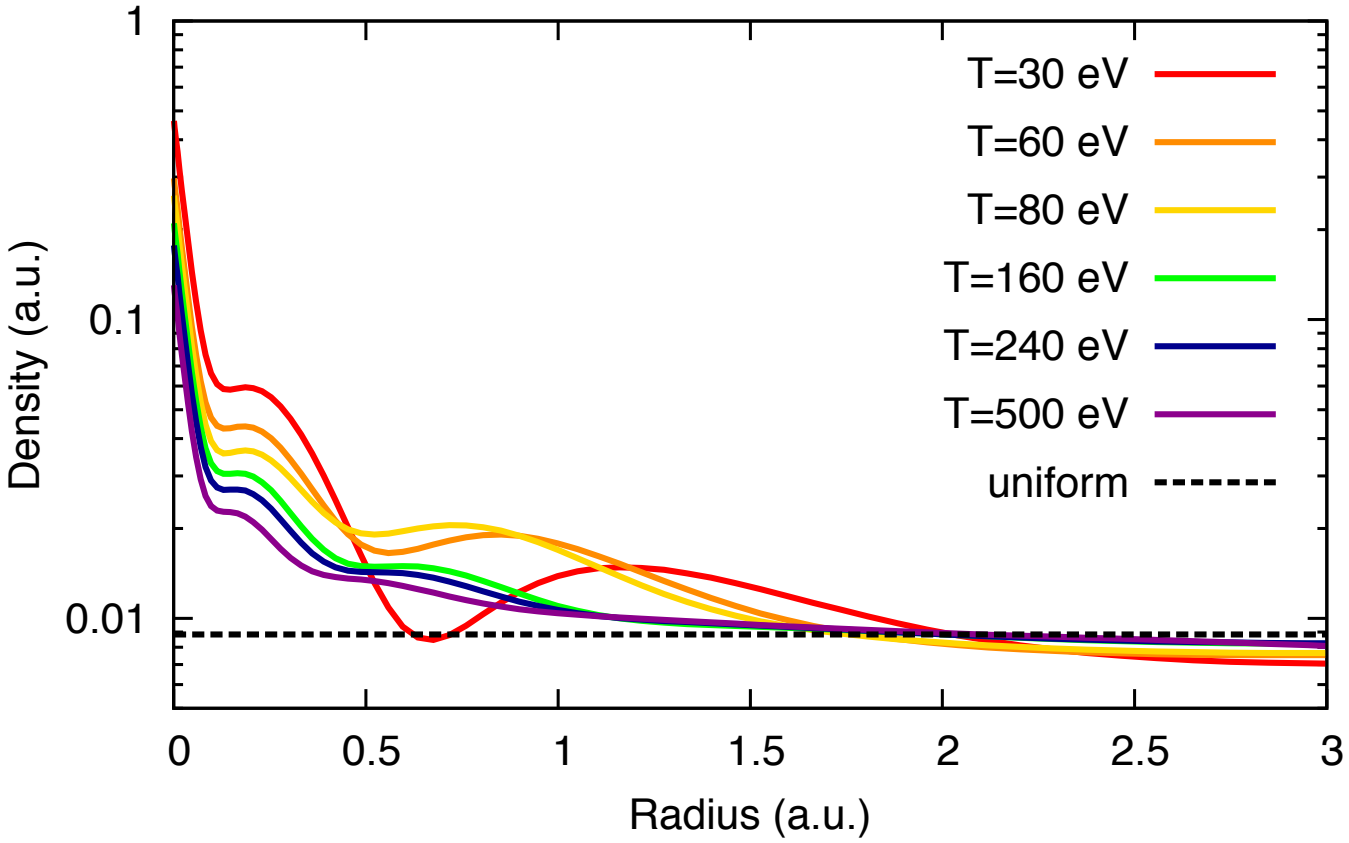
Pei & Chang, *JQSRT* **64**, 15 (2000).

First step: charge-state distribution



$$P_{[n_b]} = \prod_{b}^{\text{bound}} \frac{e^{-(\varepsilon_b-\mu)n_b/T}}{1+e^{-(\varepsilon_b-\mu)/T}} \Rightarrow P_Q = \sum_{[n_b]}^{Q} P_{[n_b]}$$

First step: free-electron density



c.f. Vinko's talk and *Nat. Commun.* **5**, 3533 (2014).

After averaging all possible free-electron configurations

$$\rho_f(\mathbf{r}, T) = \sum_{p}^{\text{continuum}} |\psi_p(\mathbf{r})|^2 \, \bar{n}_p(\mu, T)$$

Second step: fixed-config. calculation

- Connection between first step and second step
  - picking up one bound-electron configuration: most probable one
  - constructing a free-electron density
- Performing a HFS calculation

$$\hat{H} = -\frac{1}{2}\nabla^2 + V\left[\rho(\mathbf{r};T)\right]$$

$$\rho(\mathbf{r};T) = \sum_{b}^{\text{bound}} \left|\psi_b(\mathbf{r})\right|^2 n_b + \sum_{p}^{\text{continuum}} \left|\psi_p(\mathbf{r})\right|^2 \bar{n}_p(\mu,T)$$

self-consistently updated

fixed

Two-step: *K*-shell ionization & transition *E*

Al at T=80 eV

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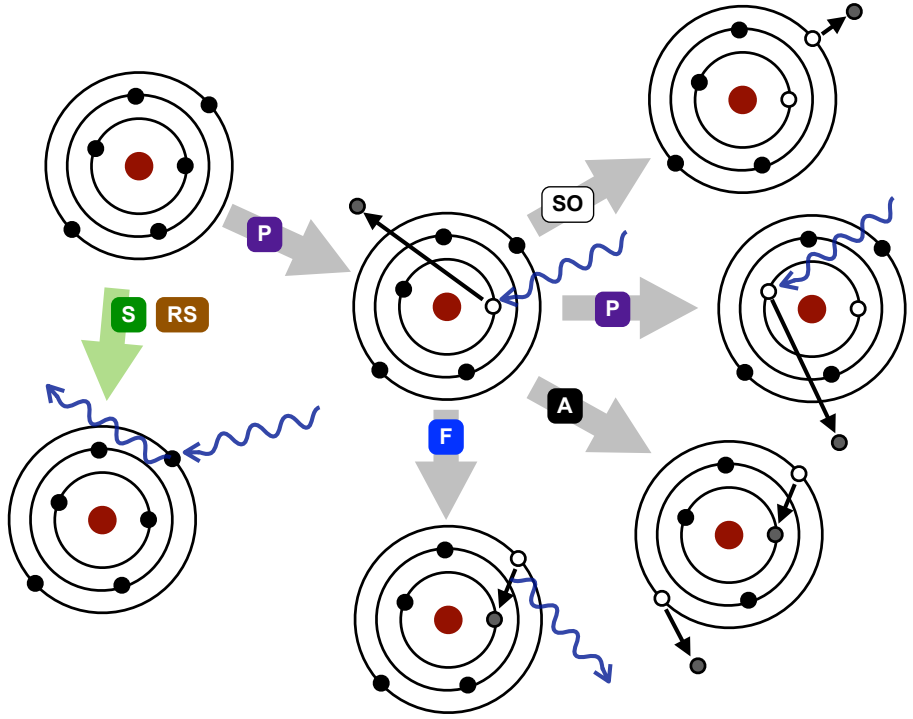
- SCF calculation for each config.
- individual configurations: different IPs and *K*α lines
- ground-state configuration ≠ the most probable configuration
- *M*-shells (3*s* and 3*p*) are bound
- *M*-shell electrons do not alter the *K*α lines ➔ K^nL^m labeling

c.f. Iglesias's talk and *HEDP* **12**, 5 (2014).

XATOM: all about x-ray atomic physics

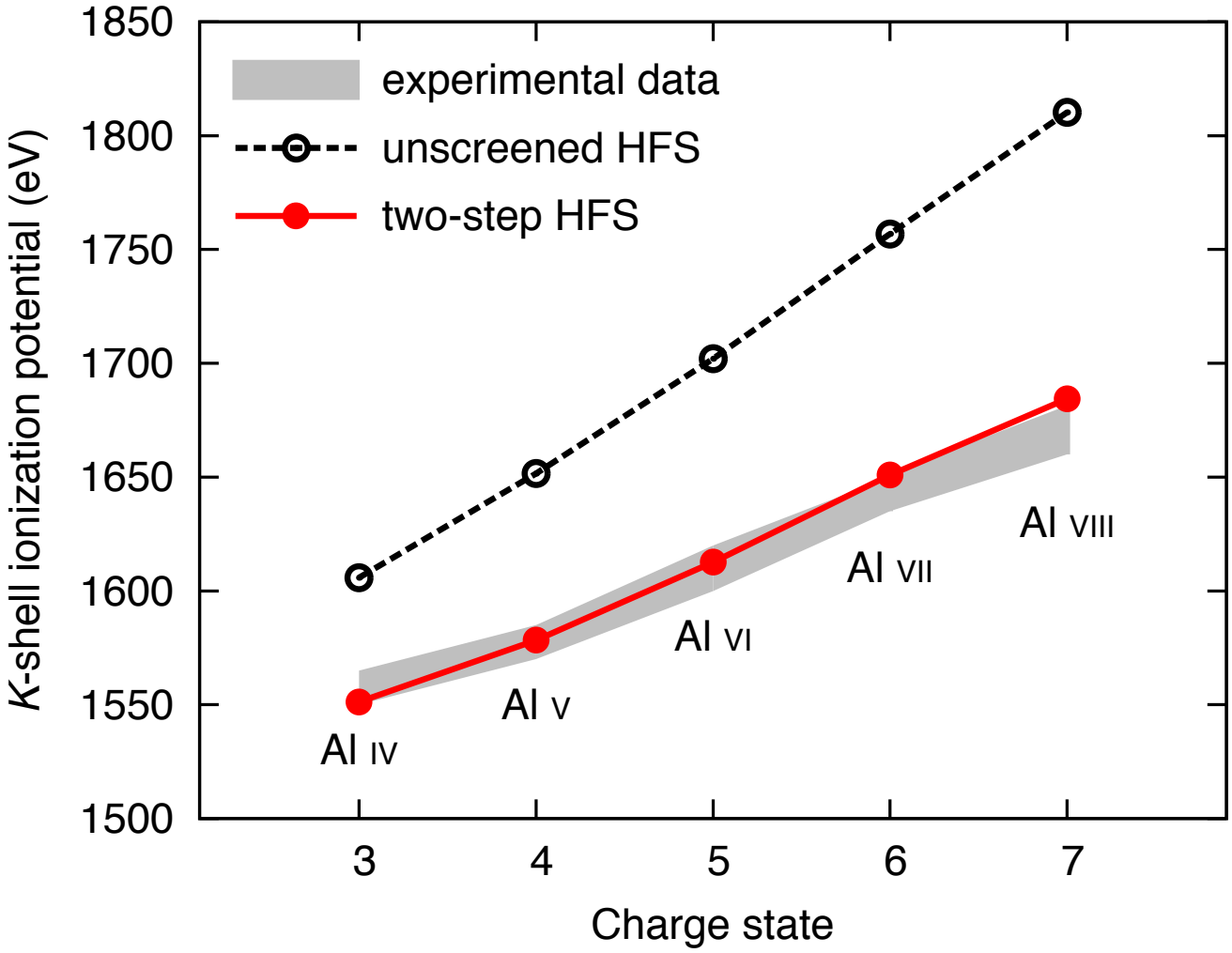
- Computer program suite to describe dynamical behaviors of atoms interacting with XFEL pulses
- Calculate all cross sections and rates of x-ray-induced processes for any given element
- Solve coupled rate equations: direct propagation or Monte-Carlo
- Well-tested for XFEL–atom experiments
- Extended to XFEL–plasma applications



Son, Young & Santra,
Phys. Rev. A **83**, 033402 (2011).

Son & Santra,
Phys. Rev. A **85**, 063415 (2012).

Comparison with LCLS experiment

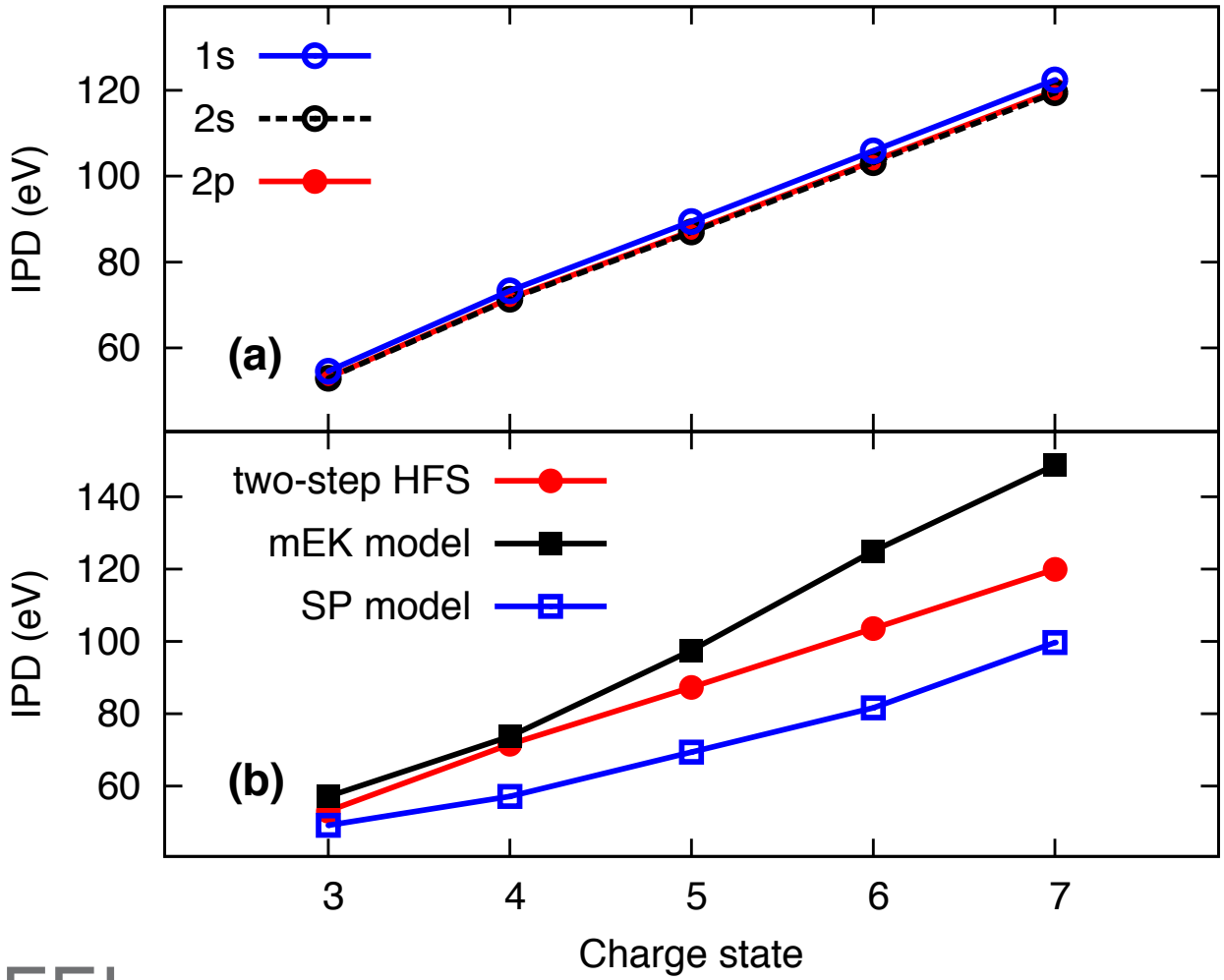


Al at T=10~80 eV
ρ=2.7 g/cm^3

Son *et al.*,
Phys. Rev. X **4**,
031004 (2014).

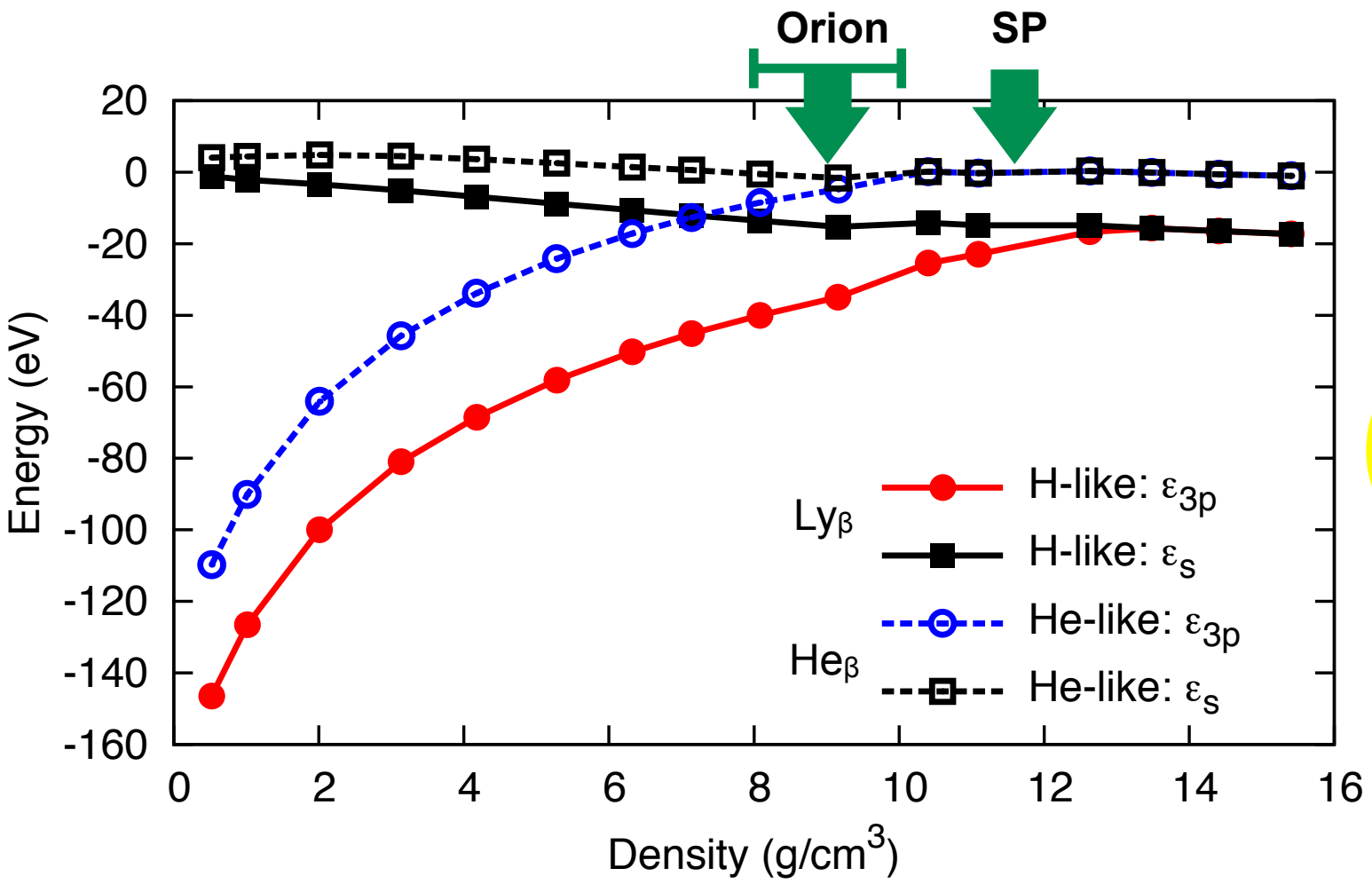
Comparison with IPD models

consistent treatment: $IPD(HFS) = IP_{atom}(HFS) - IP_{plasma}(HFS)$



Son *et al.*, *Phys. Rev. X* **4**, 031004 (2014).

Comparison with Orion experiment



Al at
T=700 eV

Son *et al.*, *Phys. Rev. X* **4**, 031004 (2014).

Conclusion

- Two-step model: first-principle calculation combining average-atom model and fixed-configuration model
- Accurately and efficiently describes atomic properties within plasmas, covering both strongly and weakly coupled plasma regimes
- Focused on IPD description of Al plasmas
- Good agreement with both LCLS and Orion experiments
- Calculated IPDs lie between SP and EK models
- Our model can be a useful tool for calculating atomic properties within dense plasmas with potentially wide-ranging applications.