An R-matrix approach to the single photon ionization of atoms and ions

Part 1 of photoionisation tutorial

Connor Ballance
(Queen's University of Belfast)

Collaborators : C Ramsbottom, F Delahaye and N Badnell

Queen's University /Strathclyde University/ Observatiore de Paris

Atomic Processes in Plasmas (May 16th 2023)

Funded in part by UK STFC, ERC Synergy grants to Queen's University of Belfast

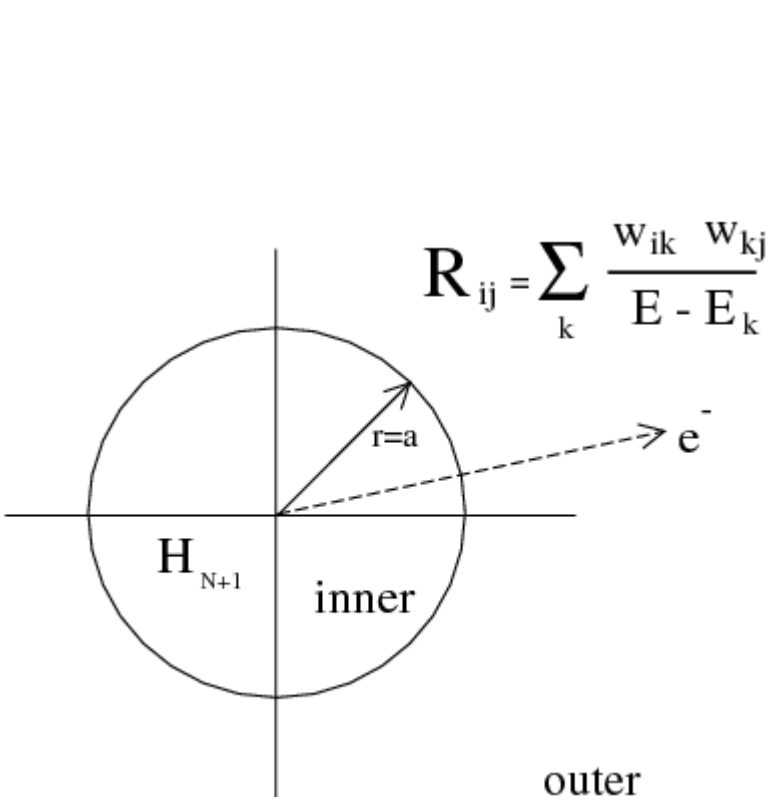
Overview

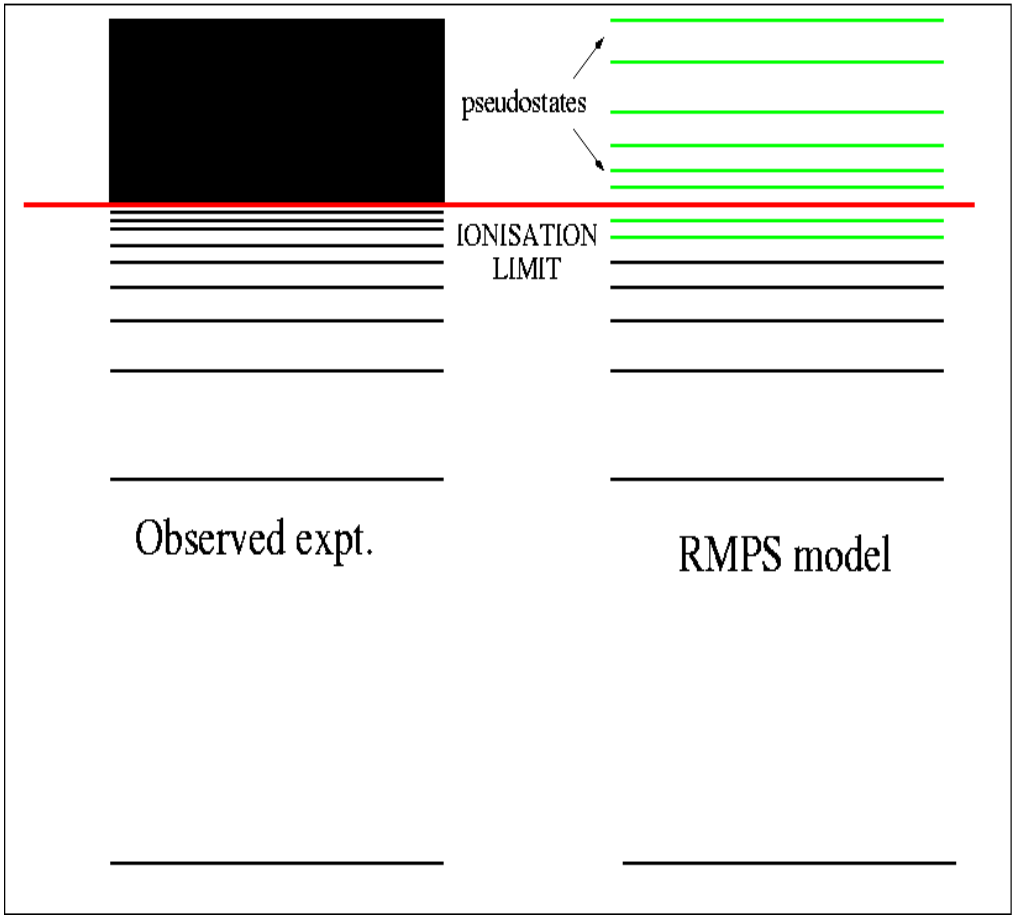
- Introduction of basic photoionization/R-matrix terminology
- Photoionization of neutral hydrogen using a configuration average approach
- Tutorial : Photoionization of He-like Fe (hv + 1s2s)
  codes + input decks : web address
  (hopefully we get this far !!)
- Discussion of the resonance features inherent within R-matrix
- Some application : comparison with experiment
  : opacity calculations (Dr Fontes : next part)

Terminology

- Terms : LS coupling representation of the target defined by spin multiplicity, angular momentum and parity (Russell-Saunders notation)
- Levels : Target states are described in J Pi notation
- R-matrix / RMPS (see next slide) : non-perturbative
- DW ,plane-wave Born, Configuration average : perturbative
- Lotz Formula , Gaunt Factors : semi-empirical
- Partial Wave : Target terms/levels couple the angular momentum of a continuum electron to give an overall N+1 sytem with specific spin, angular momentum + parity
- Scattering approaches fall into two general approaches
  Perturbative : Plane-Wave Born, Distorted-Wave
  Non-perturbative : CCC (Convergent Close coupling), R-matrix, TDCC (Time Dependent Close Coupling)

R-matrix/RMPS in a nutshell





$$\Psi_k(x_1 \ldots x_{N+1}) = A \sum_{ij} c_{ijk} \bar{\Phi}_i(x_1 \ldots x_N, \hat{r}_{N+1}\sigma_{N+1}) u_{ij}(r_{N+1}) + \sum_j d_{jk} \phi_j(x_1 \ldots x_{N+1})$$

Configuration-Average: hv + H(1s) photoionization (because a single active electron is easier to consider)

$$h\mu + (n_i l_i)^{w_i} \rightarrow (n_i l_l)^{w_i - 1} + (k_f l_f)$$

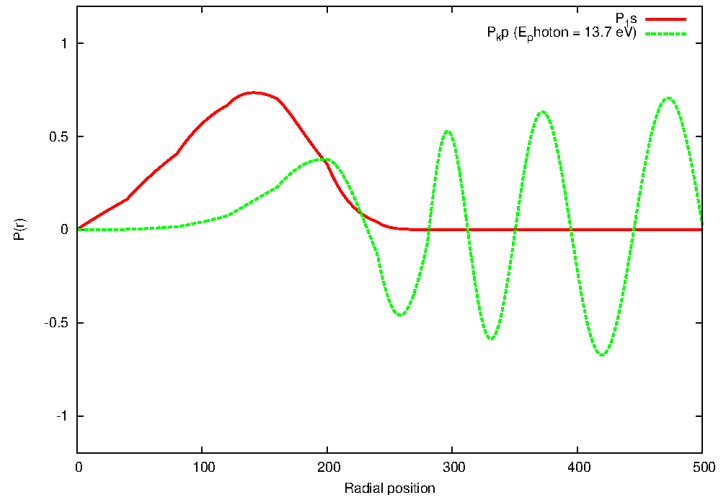
CA approx

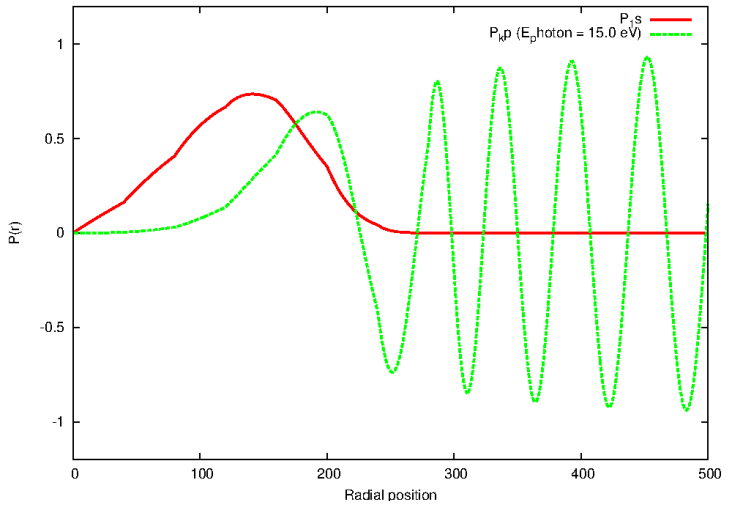
$$i.e.\ \ h\mu + (1s)^1 \rightarrow (1s)^0 + (k_f kp)$$

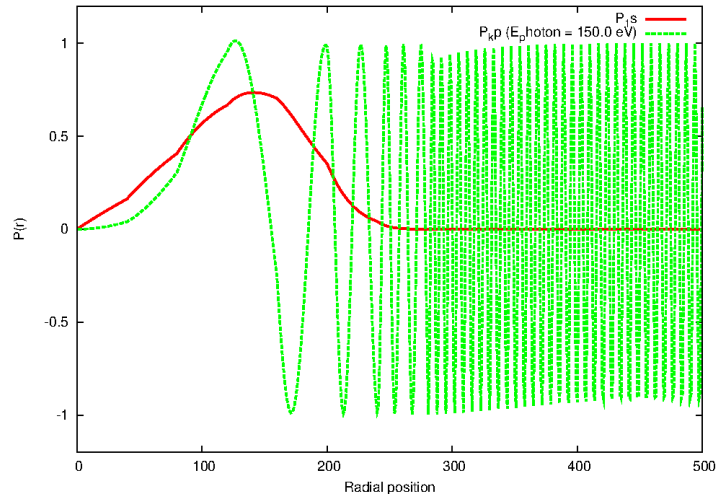
Compare with Yuri's Tutorial.

$$\sigma_{avg} = \frac{16\pi w}{3ck} \frac{w_i l_>}{4l_i + 2} \left| \int_0^\infty P_{kl}(r) r P_{n_i l_i}(r)\ dr \right|^2$$

- where w = frequency and k = electron momentum. For our particular case $l_> = 1$, $n_i = 1, l_i = 0$
- $w = I_p + k^2/2$ where I_p is the ionization potential
- atomic units are used throughout, unless stated otherwise.





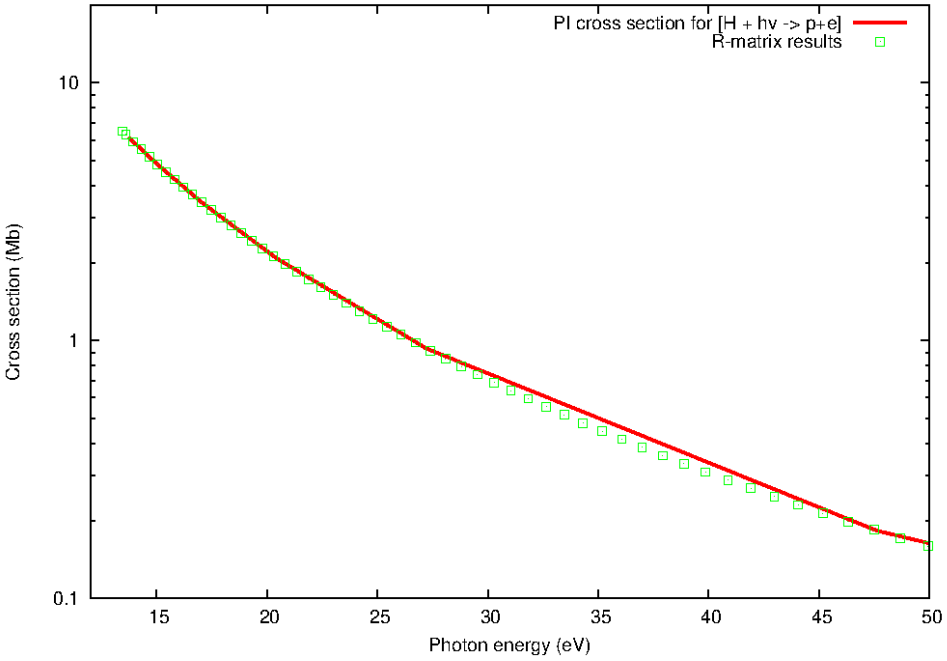


Here the 1s (red) orbital has been multiplied by r. The 3 graphs from top to bottom correspond to an incoming photon of 13.6, 15.0 and 150.0 eV (green) .

The wavefunction of representing the continuum (green) is distorted or perturbed close to the nucleus (radial position=0)

Each of plots to the left contributes a single energy point to a total photoionization plot (see below)

As the photoionisation is a mediated overlap between an initial discrete bound state and a continuum (free) wavefunction it is called a bound-free transition.



R-matrix Tutorial : photoionisation of He-like (1s2s) Fe state

Where can I get the codes used in this tutorial ?

* Codes and this particular example are available at:

  http://connorb.freeshell.org/gasp/vienna2023/ (no password required)

* There are 3 downloads: Atomic structure, Rmatrix_inner, Rmatrix_outer

  Create 3 separate directories , 'tar -xvof *.tar' in each

* 'go_compile' (perhaps after chmod u+x) & 'go_run' are scripts compile and run the codes in each section

* Works with gfortran + openmpi (therefore free). Costs only your time and a little of your sanity. This problem will run in under 2 mins on an old laptop.

* You can run these at your own leisure, I will be showing only the results from this test case, we only have a 25 min time slot.

(show usage)

R-matrix Tutorial : photoionisation of He-like (1s2s) Fe state

How do I compile and run the codes ?

Step 1: Run the atomic structure to optimise the orbitals for the structure Of the residual ion (i.e. in this case He +)

./go_compile, ./go_run

File Edit Search Preferences Shell Macro Windows Help

```
 GRASP: Fe XXVI Structure
 6 6 0  ! CSFs,orbitals,IOP,ATW
 1S  1 0 0 0 0 0
 2S  0 1 0 0 0 0
 2P  0 0 1 0 0 0
 3S  0 0 0 1 0 0
 3P  0 0 0 0 1 0
 3D  0 0 0 0 0 1
ANG  7 10
-1                !all poss Jp
MCP
MCBP
MCT  1  0         !E1 only
MCDF
0 0 0             !write new MCDF.DAT
26
FIX 0
EAL 24
BENA 20

OSCL 20 9 10 11

STOP
```

6 non-relativistic configurations 1s-3d that will be converted To relativistic orbitals within GRASP0

Solves the Coulomb-Dirac Hamiltonian by default.

Orbitals → TARGET.INP

R-matrix Tutorial : photoionisation of He-like (1s2s) Fe state

Step 2: Run the R-matrix inner region, need a continuum basis, build an N and (N+1) eelectron Hamiltonian or partial wave. Diagonalize the N+1 Hamiltonian to calculate the R-matrix on the inner region boundary.

./go_compile, ./go_run

File Edit Search Preferences Shell Macro Windows Help

```
DSTG2: DARC      ! num proc =num dipoles * np_per_sub (from dstgdto3)
  &PREINP NPW=6 NP_PER_SUBWORLD=2 IDIMCHECK=-1 IANGULAR=-1 /
  &DSTG2 NWM=6 NMAN=6 IPOLPH=2 INAST=6 /
  &ORB PRINC=1 KAPPA=-1 CSF=1 0 0 0 0 0/
  &ORB PRINC=2 KAPPA=-1 CSF=0 1 0 0 0 0/
  &ORB PRINC=2 KAPPA=-2 CSF=0 0 1 0 0 0/
  &ORB PRINC=3 KAPPA=-1 CSF=0 0 0 1 0 0/
  &ORB PRINC=3 KAPPA=-2 CSF=0 0 0 0 1 0/
  &ORB PRINC=3 KAPPA=-3 CSF=0 0 0 0 0 1/
 &ANGOPT /
 &JVALUE /
 &SYM JTOT=1.0  NPTY=1 /
 &SYM JTOT=0.0  NPTY=-1 /
 &SYM JTOT=1.0  NPTY=1 /
 &SYM JTOT=1.0  NPTY=-1 /
 &SYM JTOT=1.0  NPTY=1 /
 &SYM JTOT=2.0  NPTY=-1 /
```

Looks remarkably similar to the structure input except for the listing of dipole paired partial waves

Calculates : J=1e-0o
1e-1o
1e-2p

Dipoles

(actually concurrently as this a parallel code)

R-matrix Tutorial : photoionisation of He-like (1s2s) Fe state

Step 3: Run the R-matrix outer region region, find an initial discrete boundstate (stgb) and run a fine mesh for the photoionisation cross section (needs H.DAT and DXX files from inner region)

./go_compile, ./go_run

File Edit Search Preferences Shell Macro Windows Help

```
 &STGF
       NODAMP=1 IPERT=0 IPRINT=-2 IQDT=0 IMESH=1 IOPT1=2
       IAUGER=0 NTYP1=0 NTYP2I=0 NMIN=4
       IPHOTO=9 NPIEB=1 /
 &MESH1 MXE=10000 E0=0.5 EINCR=0.00008 &END
 0 0 1
 0 2 1
 0 4 1
-1 -1 -1
```

If we go back to our trial wavefunction ….

$$\Psi_k(x_1 \ldots x_{N+1}) = A \sum_{ij} c_{ijk} \bar{\Phi}_i(x_1 \ldots x_N, \hat{r}_{N+1}\sigma_{N+1}) u_{ij}(r_{N+1}) + \sum_j d_{jk}\phi_j(x_1 \ldots x_{N+1})$$

We know that our initial bound state (1s2s Triplet S (J=1) even) , by dipole selection rules will have 3 possible free partial waves namely, J=0 odd, 1 odd and 2 odd.

In a simplified interpretation of the above equation

Our initial state (1s2s) Triplet S (J=1) = 1s+ks, 2s+ks, …, 3d+kd (first summation)

+ 1s2s,1s3s, …, 2s3s, 2p3p (second sum)

(not all listed)

The eigenvalues of the second summation span from below the ground-state of the hydrogen like to energies above. In the former case they form the initial bound-state and in the later they may form the low members of Rydberg series seen in the photoionisation cross section.

Very brief review of input

* GRASP (General Relativistic Atomic Structure Package) – code aimed at the 100,000 level calculation that you run on your laptop

6 orbitals → 9 J Pi levels included in photoionisation calculation
R-matrix focuses on the structure of the residual ion

* stg1d_orb, pstg1d_int, pstg2d_dip, pstg3r,pstgd (R-matrix inner region)

* stgb, stgbf0 (which produces photoionisation cross sections) (R-matrix outer region)

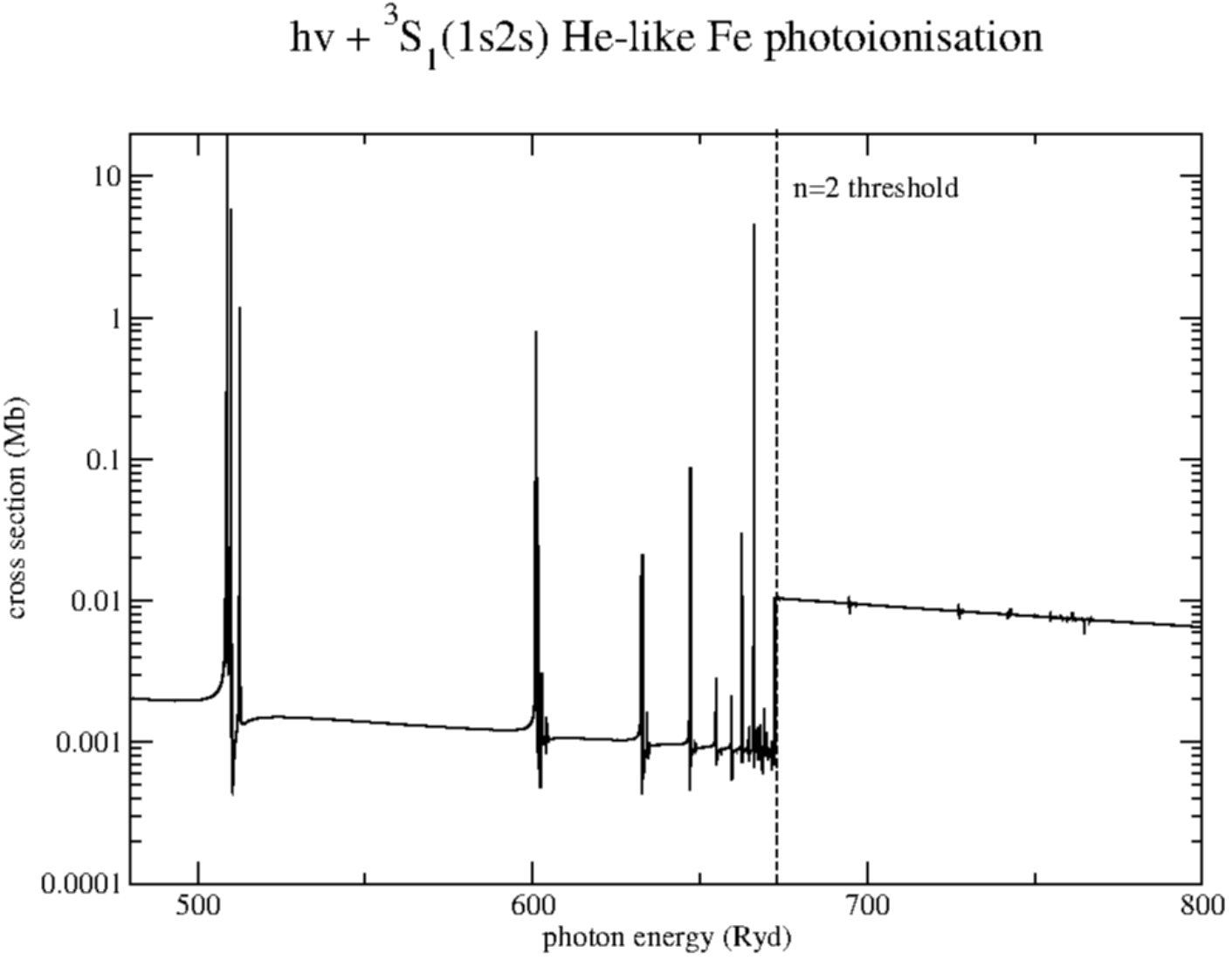
You chose how refined you wish the cross to be by the number of photon energies

(1)GRASP provides orbitals delineated on an exponential radial grid (TARGET.INP)

(2) Rmatrix_inner region provides the H.DAT (produces R-matrix) and D files (DXX) that contain the bound-free dipole matrix elements

(3) R-matrix_outer_region provides the initial bound states (stgb) and the photoioisation cross sections (pstgbf0)

Results


hv + $^3S_1$(1s2s) He-like Fe photoionisation
n=2 threshold
cross section (Mb)
10
1
0.1
0.01
0.001
0.0001
500
600
700
800
photon energy (Ryd)

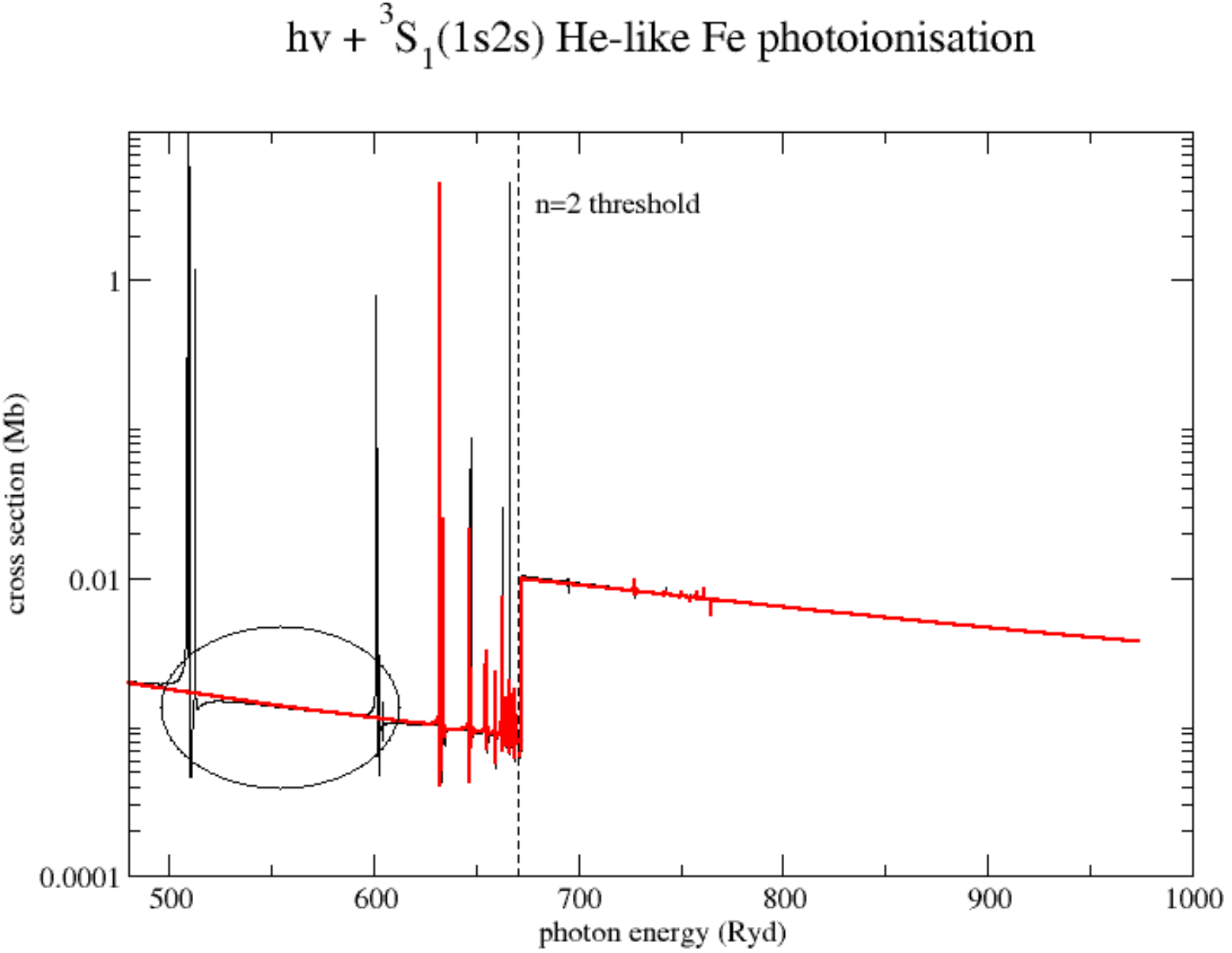
But say we remove the configurations associated with 2s,2p,3s,3p and 3d.

File Edit Search Preferences Shell Macro Windows Help

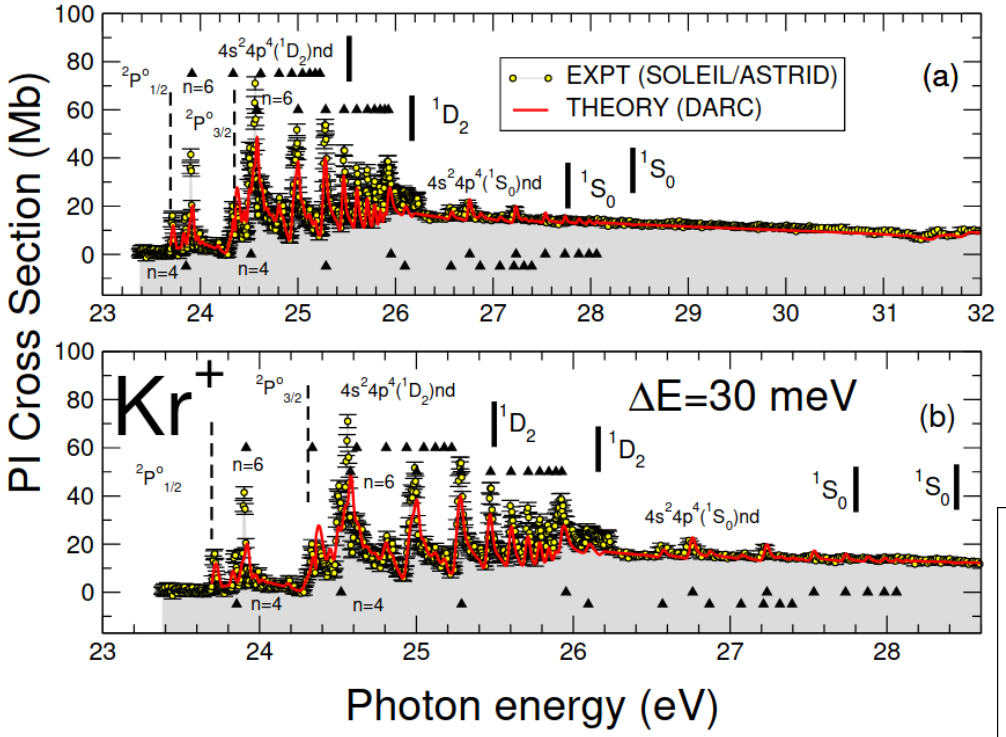
```
DSTG2: DARC       ! num proc =num dipoles * np_per_sub (from dstgdto3)
  &PREINP NPW=6 NP_PER_SUBWORLD=2 IDIMCHECK=-1 IANGULAR=-1 /
  &DSTG2 NWM=6 NMAN=6 IPOLPH=2 INAST=6 /
  &ORB PRINC=1 KAPPA=-1 CSF=1 0 0 0 0 0/
  &ORB PRINC=2 KAPPA=-1 CSF=0 1 0 0 0 0/
  &ORB PRINC=2 KAPPA=-2 CSF=0 0 1 0 0 0/
  &ORB PRINC=3 KAPPA=-1 CSF=0 0 0 1 0 0/
  &ORB PRINC=3 KAPPA=-2 CSF=0 0 0 0 1 0/
  &ORB PRINC=3 KAPPA=-3 CSF=0 0 0 0 0 1/
 &ANGOPT /
 &JVALUE /
 &SYM JTOT=1.0  NPTY=1  NCORINC=1 /
 &SYM JTOT=0.0  NPTY=-1 NCORINC=1 /
 &SYM JTOT=1.0  NPTY=1  NCORINC=1 /
 &SYM JTOT=1.0  NPTY=-1 NCORINC=1 /
 &SYM JTOT=1.0  NPTY=1  NCORINC=1 /
 &SYM JTOT=2.0  NPTY=-1 NCORINC=1 /
```

This implies only keep configurations associated with 1s target config.
i.e. remove all 2snl,2pnl,3snl,3pnl ad 3dnl where nl < 3d

Consequence : removal of first members of Rydberg sequences (ie n=2,3) or ‘in-the-box’ resonances

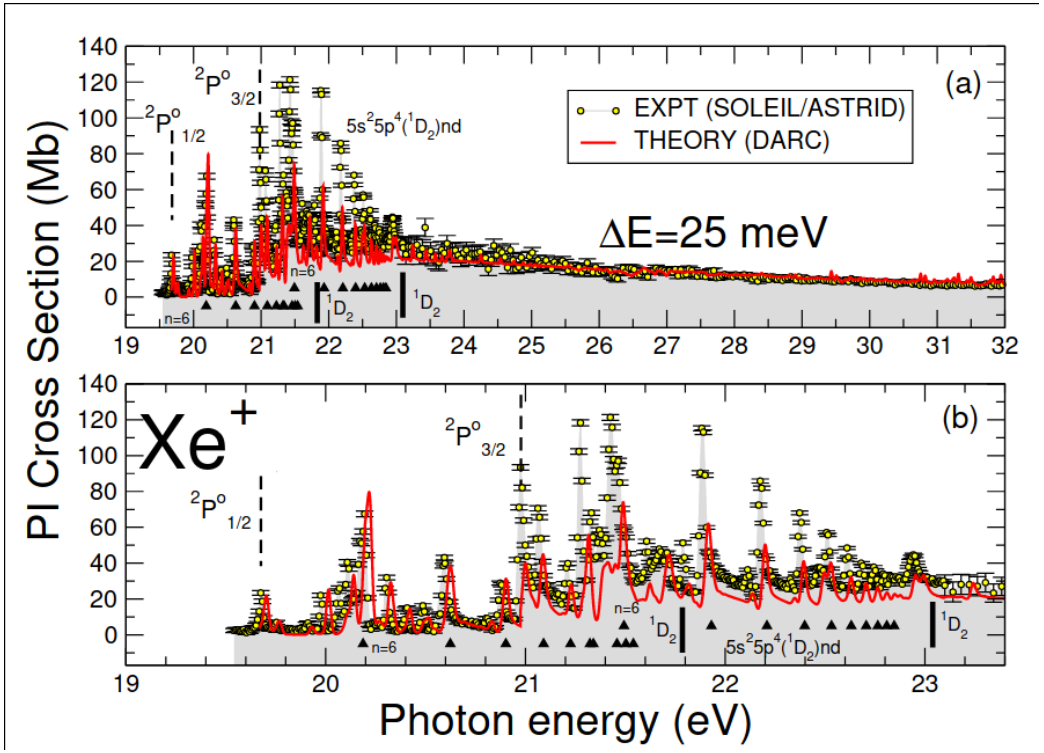


R-matrix largely reproduces the highest resolution experimental measurement



Experimental results from the SOLEIL experiment as compared to DARC theoretical values convoluted width a 25 meV width.

Resonances are all mapped out, even if the heights do not necessarily agree.



Photoionisation cross sections are one of the ingredients of opacity calculations.Although accurate,not always the most efficient approach

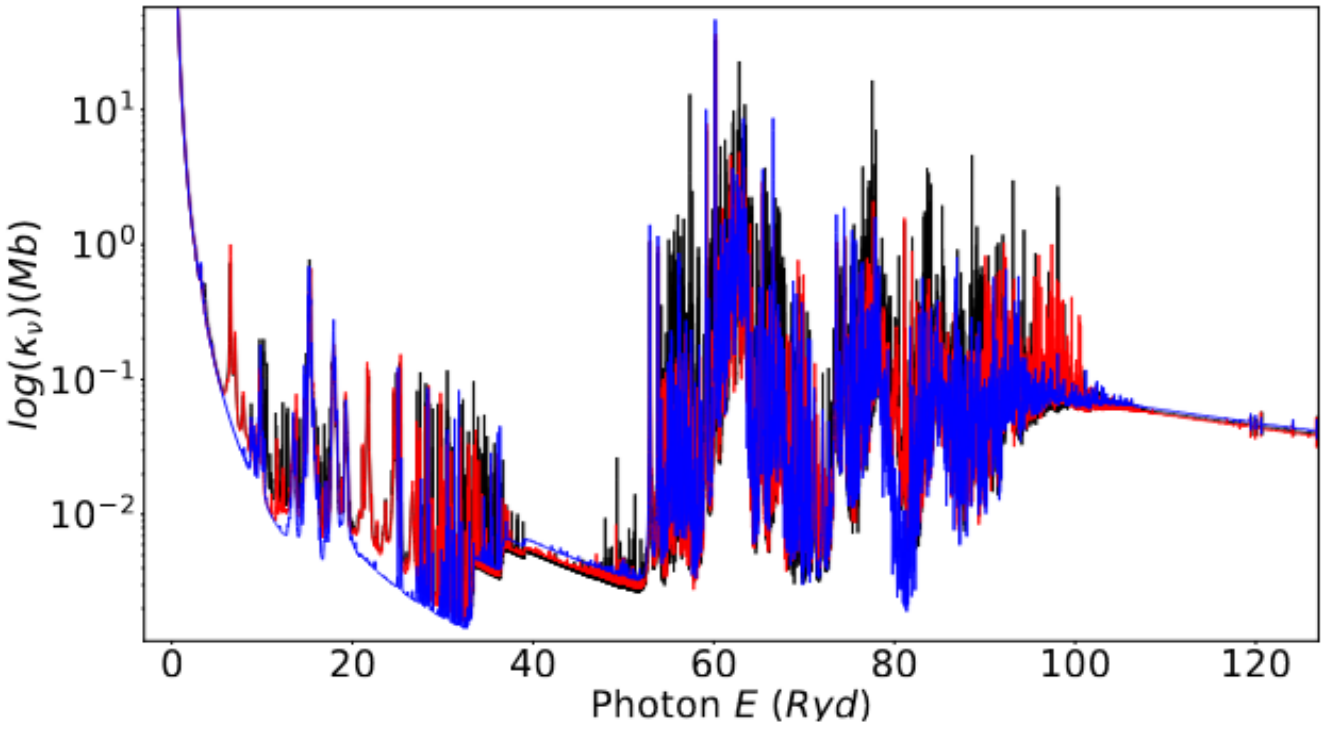


Figure 1. *R*-matrix monochromatic opacities for Fe XVII for configurations up to $n = 4$ (blue), $n = 5$ (red) and $n = 6$ (black). (Models A, B and C.)

Dr Fontes may talk about this in greater detail.