An R-matrix and AUTOSTRUCTURE approach to Dielectronic Recombination

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Some topics for discussion

- At what level of accuracy are the dielectronic recombination and radiative recombination rates required ?

  (Configuration-Averaged, S L Pi term resolved,

  J Pi level resolved)

- Do the modelling/transport codes simply require total Maxwellian DR rates , or final term/state data ?

- The first two questions are probably dependent on the ion stage and/or the particular modelling code

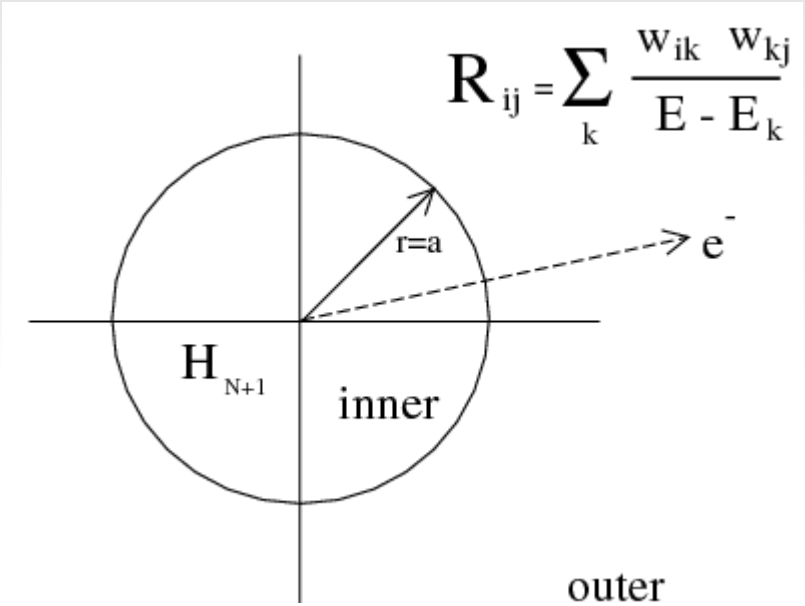
1. R-matrix and AUTOSTRUCTURE overview and comparisons.

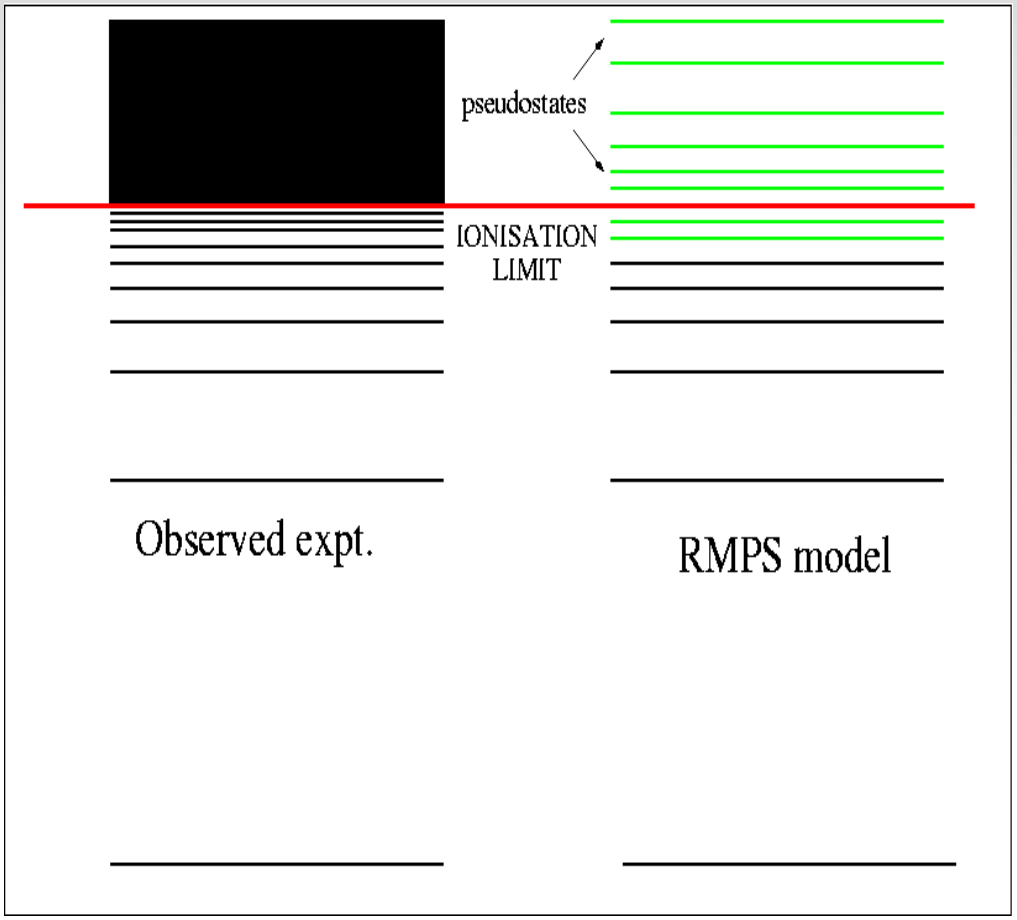
2. 'Simple' [core]1-3 electron open-shell systems (W^35+, Au^20+)

3. Multiple electron open-shell Systems(W^20+)

4. Verification through experimental Photoionisation measurements

R-matrix/R-matrix with Pseudostates (RMPS) review





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

R-matrix/R-matrix with Pseudostates (RMPS) review

Dielectronic recombination implementation is an extension of the photoionisation codes. The same 'bound-free' matrix elements for photoionisation are used for DR. i.e. Milne relationship connecting RR and photoionisation.

However you need considerably more dipole matrices for DR than photoionisation which has restrictive dipole selection rules.

Photoionisation

hv + W^{34+} 4p^64d^4

J=0 even ---> J=1 odd

A single dipole

Dielectronic Recombination

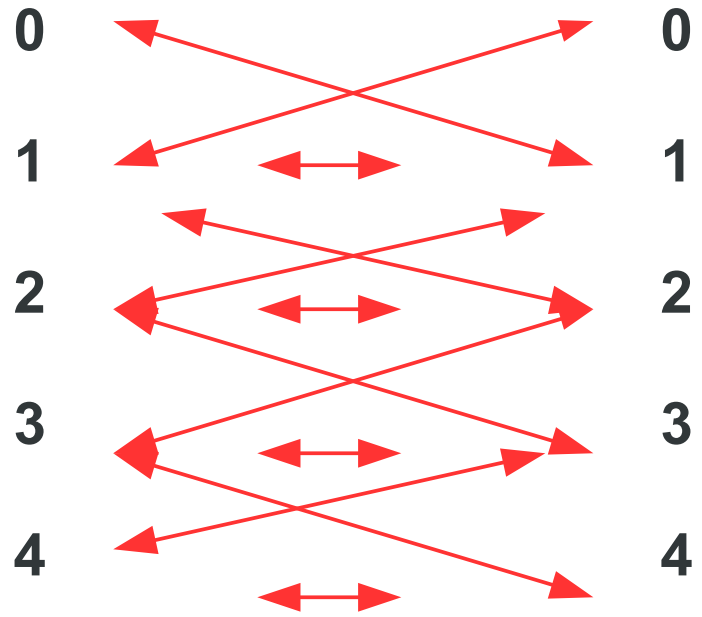
e- + W^{35+} 4d^3 → 4d^3 nl
→ 4d^2 4f nl
4d^2 5l nl
+ many more

which may radiate to hundreds/ thousands of boundstates

4d^4,4d^3 4f + may more
= many dipoles

R-matrix/R-matrix with Pseudostates (RMPS) review

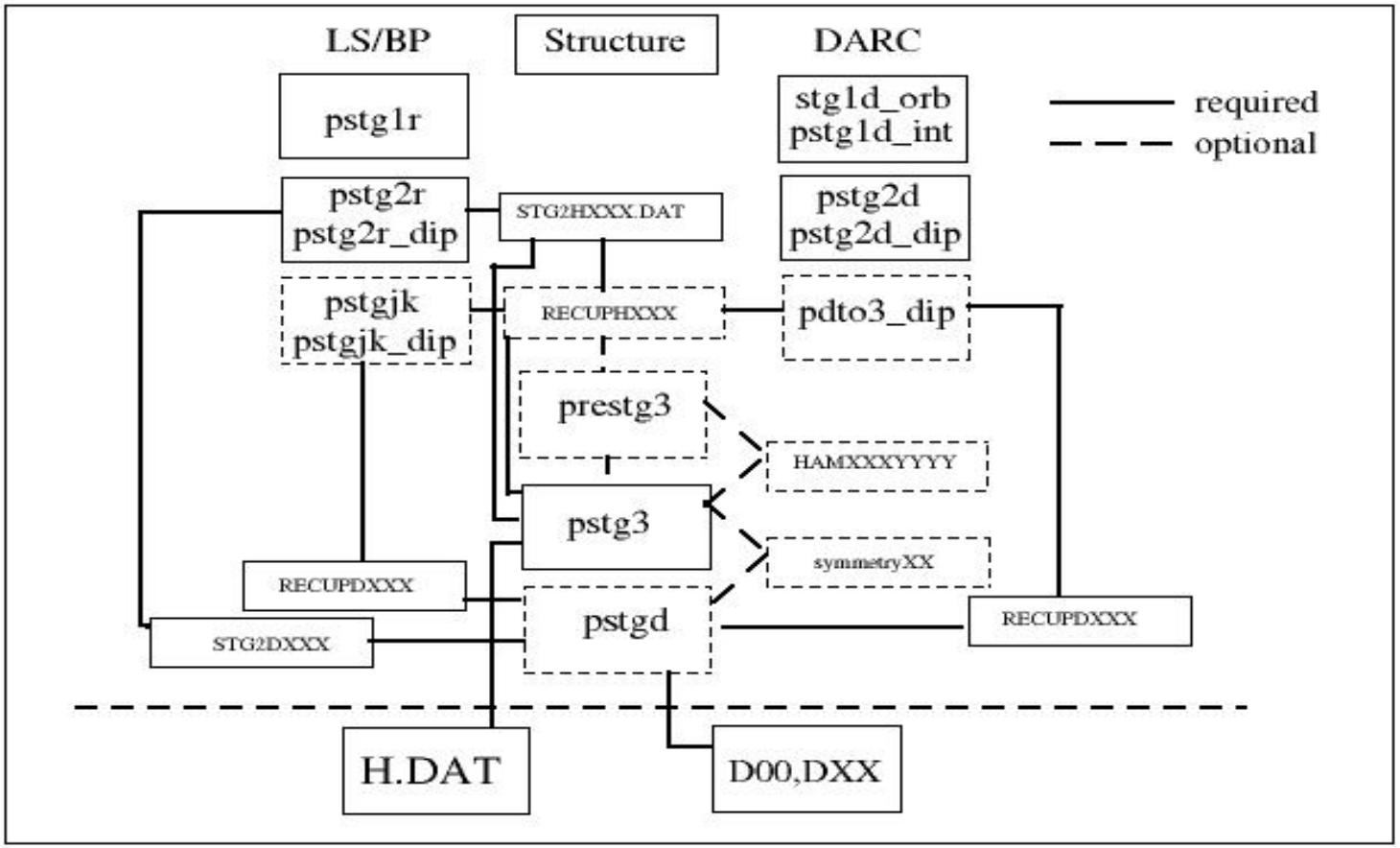
It is only through the comprehensive parallelisation of the R-matrix dipole codes that the method becomes viable for DR. It is a heavy-handed approach compared to AUTOSTRUCTURE



Every dipole pair is carried out concurrently

Each line, corresponds to a dipole pair , and an independent process

Flowchart of the R-matrix dipole codes (LS, Breit-Pauli and DARC)



AUTOSTRUCTURE

- Developed by Prof Nigel Badnell from the atomic structure code SuperStructure (Eissner, Storey and Nussbaumer 1974/1978)
- 'Auto' : the inclusion of a distorted wave to represent a continuum wavefuction, and therefore the calculation of autonisation/photoionisation
- **http://amdpp.phys.strath.ac.uk/tamoc/DATA/**

  All ionisation stages from H to Mg DR have been calculated, but in recent years autos has added more relativistics features such a the HFR (similar to the Cowan code) description of the target

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Dielectronic recombination of W^{35+}

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Benchmark comparison of the first DARC DR calculation with AUTOSTRUCTURE term/level resolved results, and DRACULA (Dielectronic Recombination Average Configuration Using the LOCAL Approximation) at the peak abundance of that ion stage

Level Resolved

$$\bar{\sigma} = \frac{2\pi^2}{\Delta\epsilon k_i^2}\frac{g_j}{2g_i}\frac{\sum_{l_i} A^a(j \to i, k_i l_i)\sum_f A^r(j \to f)}{\sum_{i',l_{i'}} A^a(j \to i', k_{i'} l_{i'}) + \sum_{f'} A^r(j \to f')}. \quad (1)$$

Configuration Averaged

$$\bar{\sigma}_{CA} = \frac{2\pi^2}{\Delta\epsilon k_i^2}\frac{G_j}{2G_i} \times \frac{\sum_{l_i} \bar{A}^a(j \to i, k_i l_i)\sum_f \bar{A}^r(j \to f)}{\sum_{i',l_{i'}} \bar{A}^a(j \to i', k_{i'} l_{i'}) + \sum_{f'} \bar{A}^r(j \to f')}, \quad (2)$$

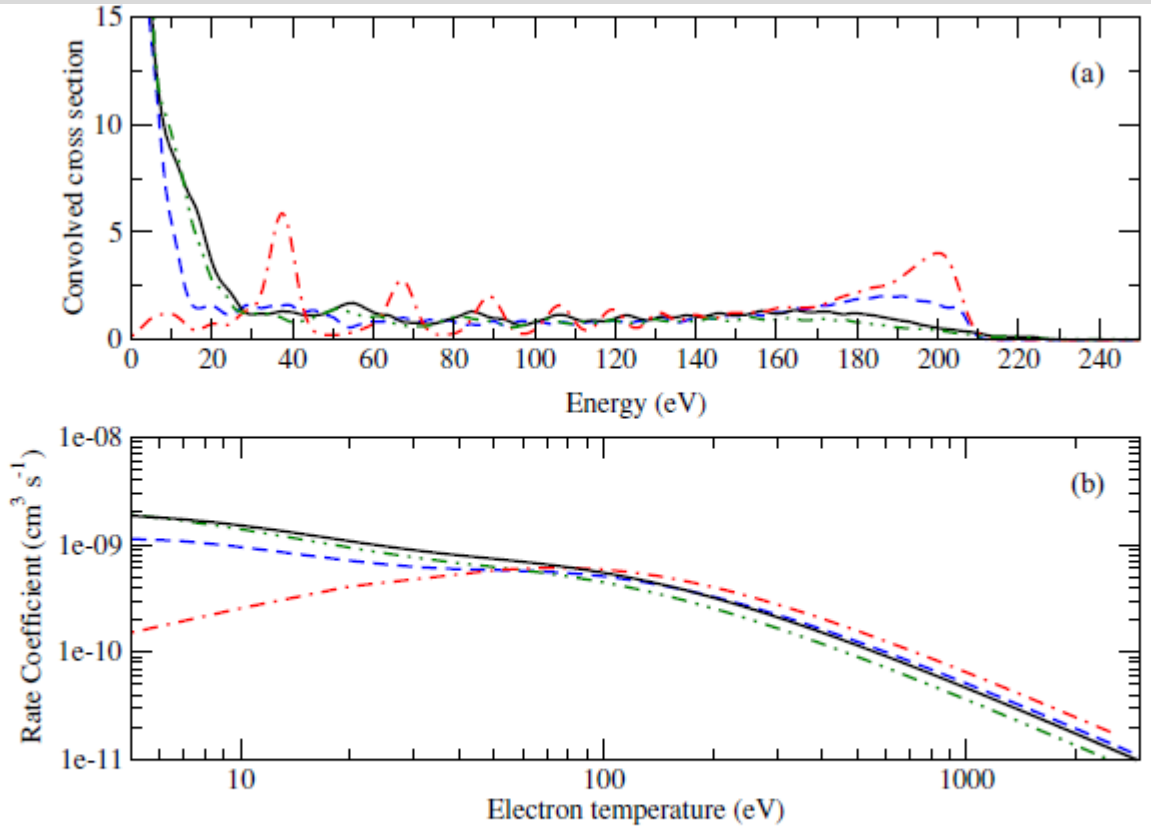

Figure 1. Dielectronic recombination (a) cross sections convoluted with a 6.8 eV Gaussian and (b) Maxwellian rate coefficients for the 4d → 4f excitation of W^{35+}. The solid curves are from the level-resolved distorted-wave calculation; the dashed curves are from the term-resolved distorted-wave calculation; the dashed double-dotted curves are from the *R*-matrix calculation; and the dot-dashed curves are from the CADW calculation.

Only the delta n=0 , or 4d-4f partial dielectronic cross section as a function of electron temeprature.

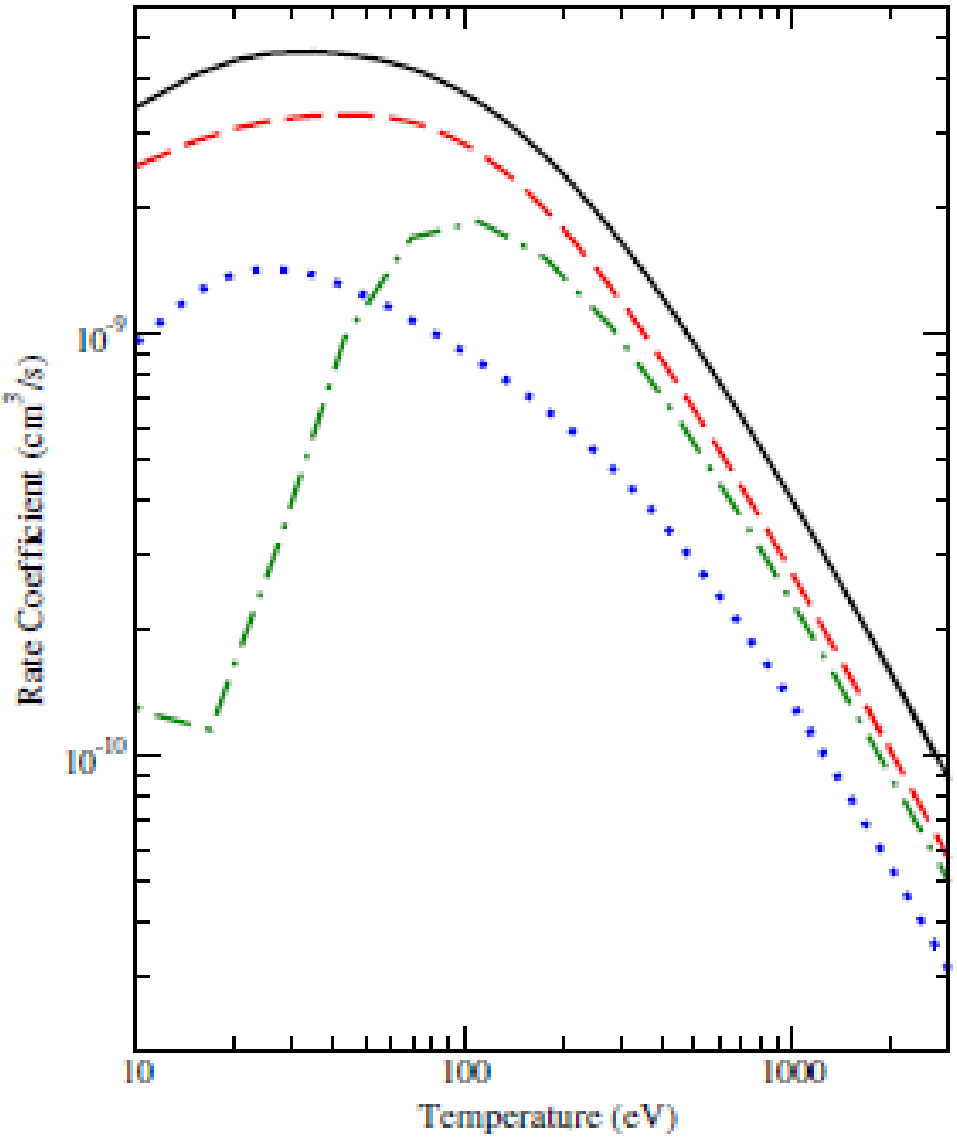

Figure 7. Total DR rate coefficients of W^{35+}. The dotted curve is the total CADW $\Delta n = 1$ rate coefficient; the dashed curve is the total CADW $\Delta n = 0$ rate coefficient; the solid curve is the total CADW rate coefficient; and the dot-dashed curve is the total rate coefficient from the average-atom approximation employed in current models.

The result confirmed earlier experiment of Thomas Putterich

Putterich T, Neu R, Dux R, Whiteford A D and
O'Mullane M G (the ASDEX Upgrade team) 2008 Plasma
Phys. Control. Fusion 50 085016

The current data (at that time) dot-dashed line had to be muliplied by 1.8 to agree with experimental Measurement. Now there is agreement.

Therefore, standard theory worked pretty well for a d^3 system and for Au^(20+) , which has a groundstate of 4f^13 we also provided good results.

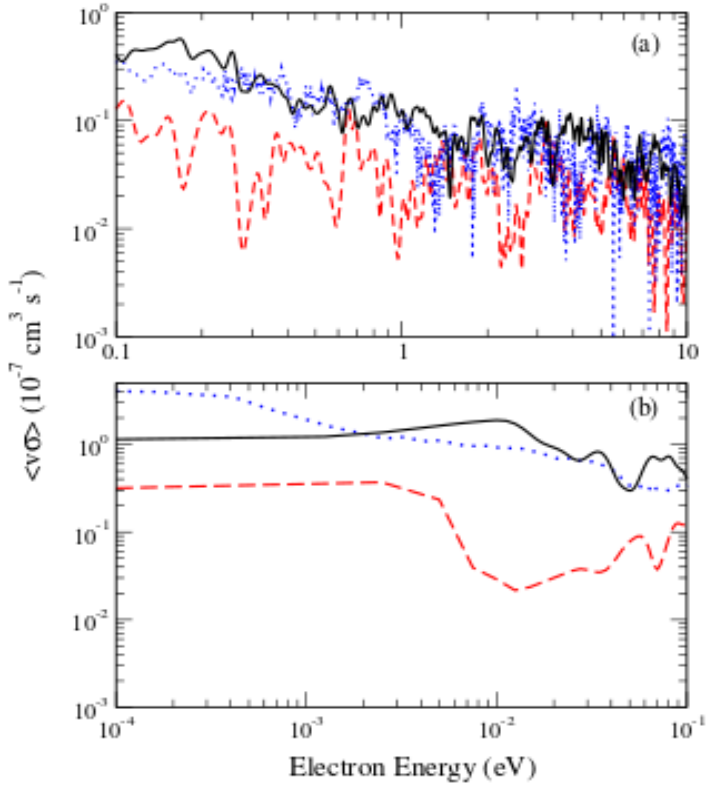

Figure 1. Dielectronic recombination rate coefficient for Au^{20+} as measured in the merged-beam experiment in (a) the higher energy range and (b) the low-energy range. The y-axis is labelled $\langle v\sigma \rangle$ to distinguish it from a Maxwellian rate coefficient. The electron-beam temperatures are $kT_{\|} = 0.1$ meV and $kT_{\perp} = 10.0$ meV. The dotted (blue) curves are from the experimental measurements of Schippers *et al* [4]; the dashed (red) curves are from the present term-resolved, *LS*-coupling calculation and the solid (black) curves are from the present level-resolved, intermediate-coupling calculation.

Level resolved cross sections as opposed to term resolved cross sections were required at the lowest temeperatures

Theoretical rate coefficients $\langle v\sigma \rangle$ were calculated using both a Maxwellian and an experimental merged-beams velocity distribution in the centre-of-mass frame of the ion, given by [12]

$$f(v_0, \mathbf{v}) = \left\{ \frac{m_e}{2\pi k T_{\|}} \right\}^{\frac{1}{2}} \exp\left(-\frac{m_e (v_{\|} - v_0)^2}{2kT_{\|}} \right) \frac{m_e}{2\pi k T_{\perp}} \times \exp\left(-\frac{m_e (v_{\perp})^2}{2kT_{\perp}} \right), \quad (2)$$

where $v_0 = \sqrt{(2E_0/m_e)}$ and E_0 is the electron–ion centre-of-mass energy; $T_{\perp}$ and $T_{\|}$ are the temperatures corresponding to motion perpendicular and parallel to the ion beam, respectively; finally, $v_{\perp}$ and $v_{\|}$ correspond to the perpendicular and parallel components of $\mathbf{v}$.

Therefore, for either 3 electrons outside a core (W^35+) or one electron away from a closed f shell (Au20+), standard perturbative theory from either DRACULA or AUTOSTRUCTURE provides good agreement with experiment.

However , W^(20+) , the agreement was not so good for standard theory

PHYSICAL REVIEW A **85**, 052716 (2012)

Dielectronic recombination of W^{20+} ($4d^{10}4f^{8}$): Addressing the half-open f shell

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(Received 9 April 2012; published 29 May 2012)

A recent measurement of the dielectronic recombination (DR) of W^{20+} [Schippers *et al.*, Phys. Rev. A **83**, 012711 (2011)] found an exceptionally large contribution from near-threshold resonances ($\lesssim$1 eV). This still affected the Maxwellian rate coefficient at much higher temperatures. The experimental result was found to be higher by a factor of 4 or more than that currently in use in the 100- to 300-eV range, which is of relevance for modeling magnetic fusion plasmas. We have carried out DR calculations with AUTOSTRUCTURE which include all significant single-electron promotions. Our intermediate-coupling (IC) results are more than a factor of 4 larger than our LS-coupling ones at 1 eV but still lie a factor of 3 below experiment here. If we assume complete (chaotic) mixing of near-threshold autoionizing states, then our results come into agreement (to within 20%) with experiment below $\lesssim$2 eV. Our total IC Maxwellian rate coefficients are 50%–30% smaller than those based on experiment over 100–300 eV.

DOI: 10.1103/PhysRevA.85.052716 PACS number(s): 34.80.Lx, 34.80.Dp

The half-open f shell, provides a severe computational challenge unless you have generous computational resources.

Before considering the incoming electron, 4f^7 / 4f^8 goundstate configurations produce well over a hundred terms and several hundred levels

To quote N R Badnell :

> The (near-) half-open f-shell problem is a daunting one. If we view it simply in terms of binomial coefficients for the number of states present in a configuration, then moving from the $4d^{10}4f^{13}$ ground configuration considered in Ref. [12] to $4d^{10}4f^{8}$ increases the number of states by a factor of $(429/2)$. Memory requirements (CPU and disk) scale as $(429/2)^2$, and the time requirement as $(429/2)^3$. This is all relative. Absolute numbers are much larger once we start promoting electrons from the $4d$ and $4f$ subshells of the ground configuration. (The only bonus of the binomial effect is that $4f^7$ is only marginally worse than $4f^8$ since the number of states increases only by a factor $8/7$.)

Without parallelism, the calculation would have been unfeasible.

However, there was an experimental measurement from Schippers et al, and for very low energies

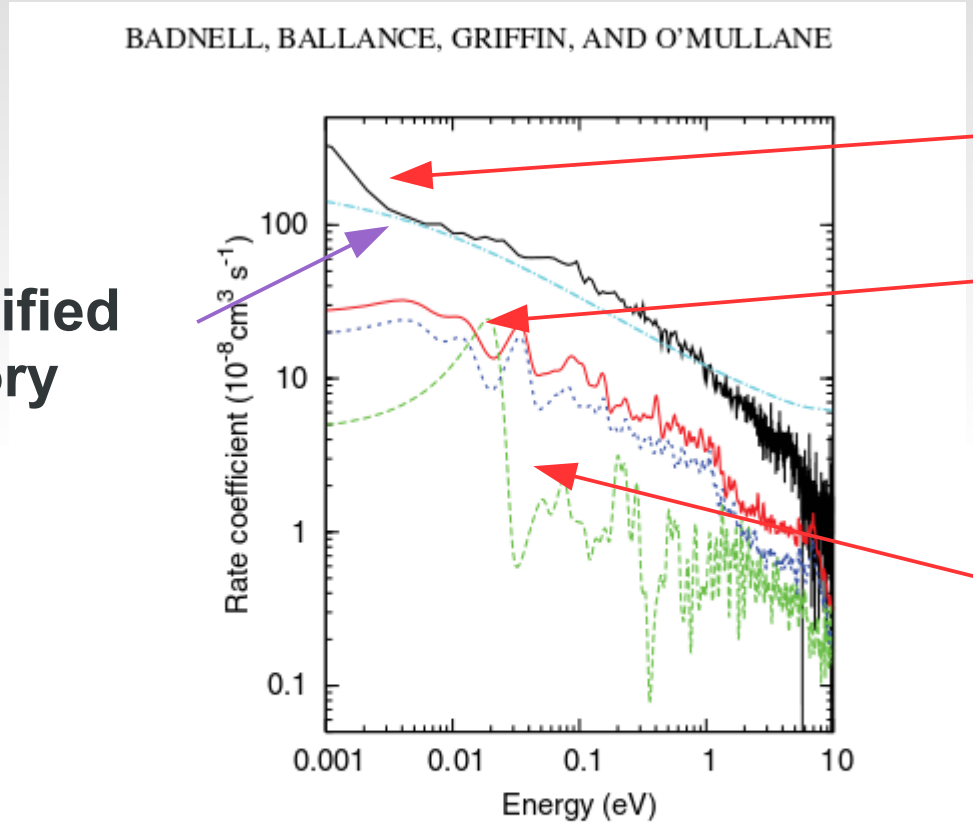

FIG. 5. (Color online) W^{20+} merged-beam DR rate coefficients: experiment [7] [upper solid (black) curve], partitioned total [dot-dashed (cyan) curve], IC total [lower solid (red) curve], *LS* total [long-dashed (green) curve], and IC $4d \rightarrow 4f$ only [short-dashed (blue) curve].

Modified theory

Experimental measurement

Intermediate coupling AUTOSTRUCTURE result

Existing data within ADAS

Therefore standard IC theory was a couple of orders of magnitude below the experimental result , at low energies

R-matrix theory still struggles with this magnitude of this calculation,

For an exploratory calculation , let us consider only the 4f^7, 4f^65s , 4f^65p , 4f^65d, 4d^54f^8 target states (~8000 levels)

level	J	parity	CSF	mix	a.u.	Ryd.
1	7/2	odd	222	0.515	0.00000000E+00	0.00000000E+00
2	7/2	odd	237	0.672	2.96695081E-01	5.93390161E-01
3	5/2	odd	277	0.482	3.21306284E-01	6.42612568E-01
4	7/2	odd	235	0.458	3.39695020E-01	6.79390040E-01
5	3/2	odd	301	0.577	3.40710657E-01	6.81421314E-01
6	9/2	odd	185	0.426	3.56290116E-01	7.12580232E-01
7	17/2	odd	38	0.505	3.66959346E-01	7.33918693E-01
8	11/2	odd	129	0.463	3.70560363E-01	7.41120726E-01
9	13/2	odd	90	0.550	3.78160966E-01	7.56321932E-01
10	15/2	odd	57	0.568	3.79488707E-01	7.58977414E-01
11	9/2	odd	174	0.669	4.00367970E-01	8.00735939E-01
12	7/2	odd	222	0.393	4.29346653E-01	8.58693307E-01

Configuration-interaction mixing is strong , more so for Rydberg states

One possible solution to the problem has been offered by the following paper, which corresponds to the dot-dashed curve of the previous page.

PHYSICAL REVIEW A VOLUME 50, NUMBER 1 JULY 1994

Structure of compound states in the chaotic spectrum of the Ce atom: Localization properties, matrix elements, and enhancement of weak perturbations

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(Received 23 December 1993)

The aim of the present paper is to analyze a realistic model of a quantum chaotic system: the spectrum and the eigenstates of the rare-earth atom of Ce. Using the relativistic configuration-interaction method the spectra and the wave functions of odd and even levels of Ce with $J = 4$ are calculated. It is shown that the structure of the excited states at excitation energies above 1 eV becomes similar to that of the compound states in heavy nuclei. The wave functions of the excited states are chaotic superpositions of the simple basis states (with the number of "principal" components $N \sim 100$), built of the $4f$, $6s$, $5d$, and $6p$ single-electron orbitals. The localization of the eigenstates on the energy scale is characterized by the spread width $\Gamma \sim ND$, where D is the average level spacing ($D \sim 0.03$ eV). The emergence of chaos in the spectrum and the dependence of the N and Γ parameters on the excitation energy are studied. The shape of the localization is shown to be Lorenzian around the maximum (principal components), whereas outside this region the squared components display a faster decrease, in agreement with the perturbation theory treatment of the band random matrix (BRM) model. The structure of the real interaction matrix is compared with that assumed in the BRM models. A formula expressing the mean-squared values of matrix elements between the eigenstates in terms of their parameters and single-particle occupancies is derived, and its applicability is checked with the results of numerical calculations. The hypothesis of a Gaussian distribution of the eigenstates' components and matrix elements between the eigenstates has been checked. The existence of the statistical (dynamical) enhancement of weak perturbations in systems with dense spectra is demonstrated.

However, this only affects very low energy results

And more recently ….

PHYSICAL REVIEW A **88**, 062713 (2013)

Electron recombination, photoionization, and scattering via many-electron compound resonances

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Highly excited eigenstates of atoms and ions with open f shell are chaotic superpositions of thousands, or even millions, of Hartree-Fock determinant states. The interaction between dielectronic and multielectronic configurations leads to the broadening of dielectronic recombination resonances and relative enhancement of photon emission due to opening of thousands of radiative decay channels. The radiative yield is close to 100% for electron energy $\lesssim$1 eV and rapidly decreases for higher energies due to opening of many autoionization channels. The same mechanism predicts suppression of photoionization and relative enhancement of the Raman scattering. Results of our calculations of the recombination rate are in agreement with the experimental data for W^{20+} and Au^{25+}.

DOI: 10.1103/PhysRevA.88.062713 PACS number(s): 34.80.Lx, 31.10.+z, 34.10.+x

By distributing the particles among orbitals in different ways one generates the Slater determinant states $|i\rangle$ (configuration states) from some mean-field, e.g., Hartree-Fock, single-particle orbitals. These states serve as the basis for finding the *eigenstates* $|n\rangle = \sum_i C_i^{(n)}|i\rangle$. When the residual interaction between the particles exceeds the energy spacing between the basis states coupled by this interaction, the eigenstates become chaotic superpositions of thousands or even millions of basis states $|i\rangle$.

The expansion coefficients $C_i^{(n)}$ in such superpositions behave largely as independent random variables. They are, however, subject to the normalization condition $\sum_i |C_i^{(n)}|^2 = \sum_n |C_i^{(n)}|^2 = 1$. Also, the variance of $C_i^{(n)}$ displays a systematic variation with the energy of the eigenstates and basis states [1,2]:

$$\overline{|C_i^{(n)}|^2} = \frac{D}{2\pi}\frac{\Gamma_{\rm spr}}{(E_n - E_i)^2 + \Gamma_{\rm spr}^2/4}. \qquad (1)$$

Here D is the mean level spacing between the basis states (or eigenstates) with a given total angular momentum and parity J^p, and $\Gamma_{\rm spr} = 2\pi\overline{H_{ik}^2}/D$ is the *spreading width*. It is

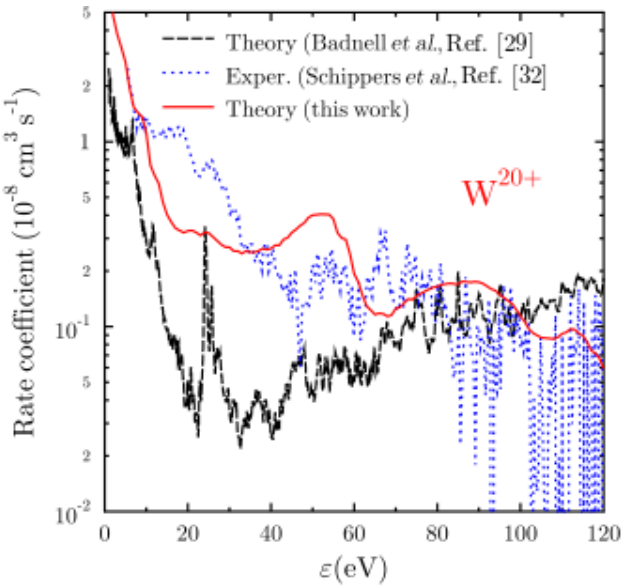

FIG. 3. (Color online) Recombination rates for W^{20+}. Solid red line is our theory, dashed black line is the calculation of Ref. [29], and dotted blue line is the experimental data [32].

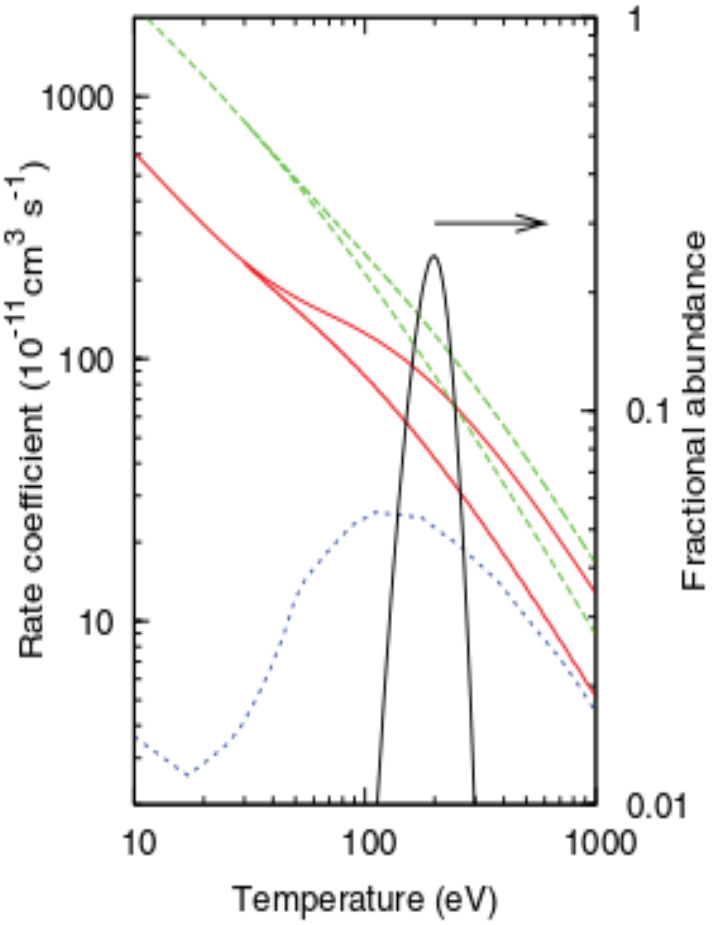

FIG. 8. (Color online) W^{20+} total Maxwellian DR rate coefficients: IC, all resonances and to 140 eV only [solid (red) curves originating on y axis]; experiment [7], to 140 eV and with theory top-up for resonances above 140 eV [long-dashed (green) curves]; and ADAS [31] [short-dashed (blue) curve]. The fractional abundance of W^{20+} in a magnetic fusion plasma is also shown [solid (black) curve originating on x axis].

In terms of modelling a magnetically confined fusion device, the large orders of magnitude difference do not necessarily translate to such large differences at the temepratures that ionisation fraction curves predict that ion stage will exist.

Our recommended data set was an amalgamation of the experimental result at low energies, with theoretical values for the high energies.

In the absence of detailed dielectronic recombination measurements for the low charge states of Tungsten, only photoionisation measurements from the ALS experiments in Berkeley provide an indication of how well we model the near-neutral stages

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Photoionization of the valence shells of the neutral tungsten atom

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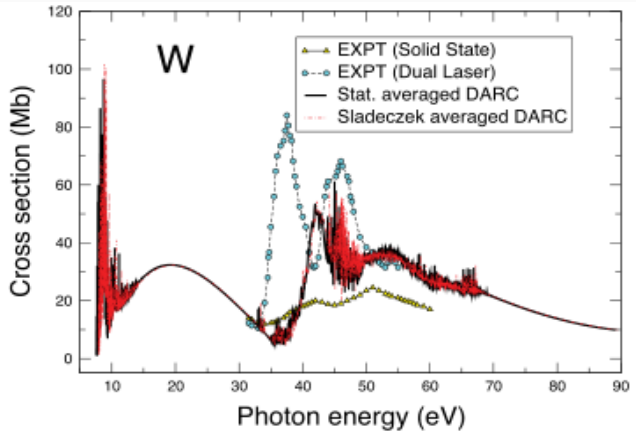

Figure 2. Single photoionization of neutral W over the photon energy range 8–100 eV, comparing weighted averaged theoretical calculations with several different experiments. The DARC results (645-levels approximation, solid black line), are the statistical average of the five levels associated the ^{5}D term ground state. The (dashed red-line) is the weighted DARC results of the lowest six levels, employing the mixing coefficients reported by Sladeczek and co-workers [6]. Solid circles with dashed line are the dual-laser experimental results of Costello and co-workers [3] and the solid triangles are the results from the experimental work of Haensel and co-workers [5]. The statistically averaged DARC PI cross sections were Gaussian convolved at a FWHM of 250 meV.

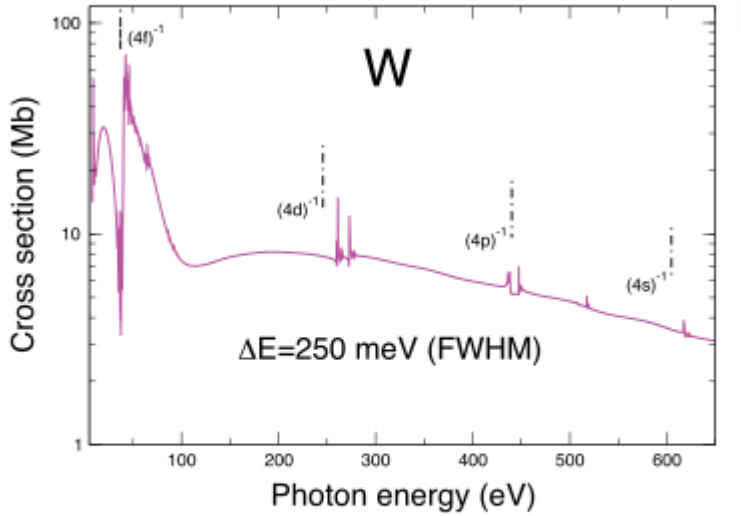

Figure 3. Single photoionization cross sections of neutral W over the photon energy range 8–625 eV, from the present DARC PI calculations incorporating 1227 levels in the close-coupling calculations. The photoionization threshold of each of the $4\ell^{-1}$ orbitals hole configurations are indicated on the graph.

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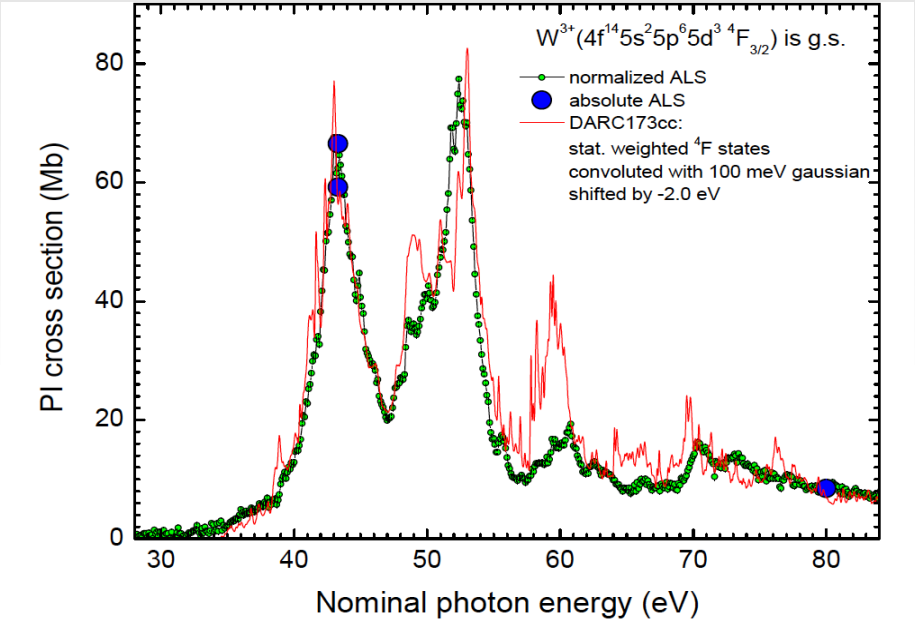
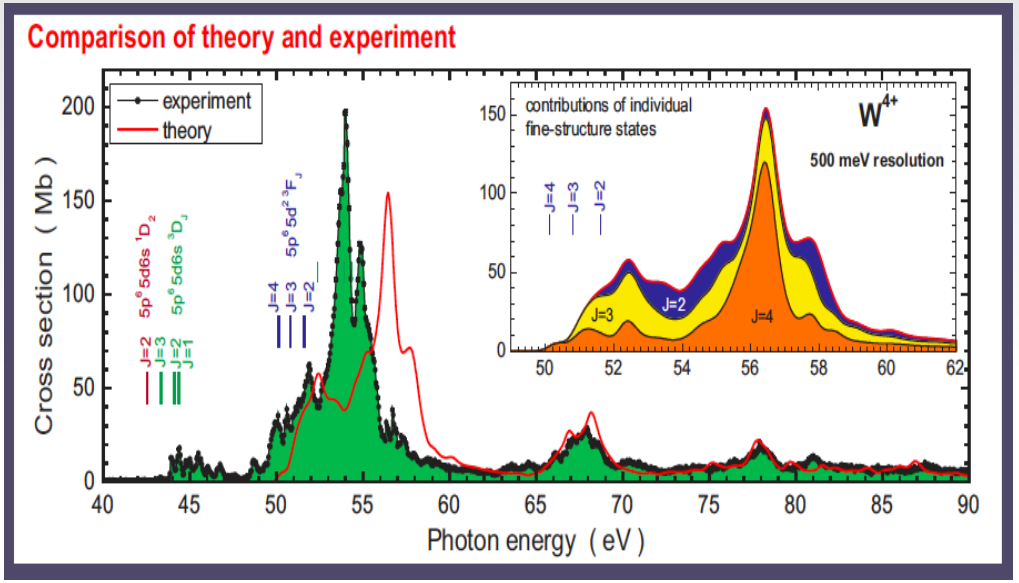
Having built 8-15 configurations models for W^+-W^4+, it seems that 300-500 level models reproduce the resonance features, with 500-900 levels improving resonance position …. **good news for electron-impact excitation!**





A. M. Mueller, S. Schippers, J. Hellhund, A. L. D. Kilcoyne, R. A. Phaneuf, C. P. Ballance and B. M. McLaughlin HCI 2012, Heidelberg, Germany (2012)

An overview of the parallel DARC dipole photoionisation codes