Atomic structure and dynamics

-- need and requirements for accurate atomic calculations

- Analysis and interpretation of optical and x-ray spectra (astro physics)
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- ......
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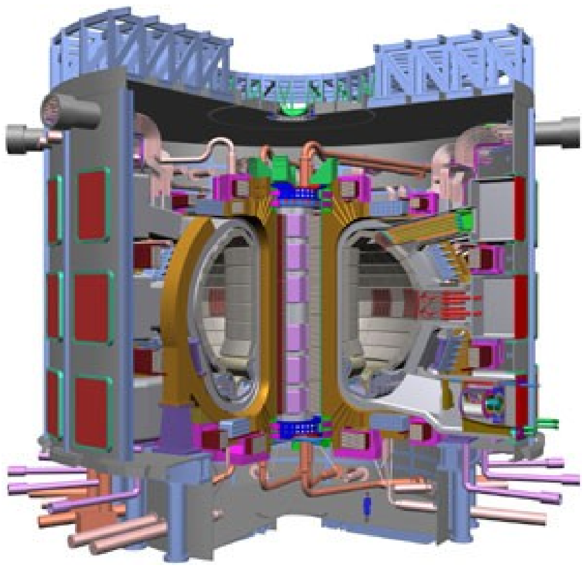


Relativistic effects in heavy and superheavy elements

... to be considered in accurate atomic calculations

S. Fritzsche, MPI-K and GSI Darmstadt

11th October 2007

Atomic interactions are well known:

- QED as the well established basis
- Atomic shell modell
  - ➔ $\psi (r,\theta,\phi) = R_{nl}(r)\, Y_{lm}(\theta,\phi)$,
  - ➔ „aufbau principle“: Successive filling of subshells and shells

Outline of this talk:

i) „Electronic correlations“: The challenge of open shells

ii) (Super-) Heavy elements: Rapid increase of relativity

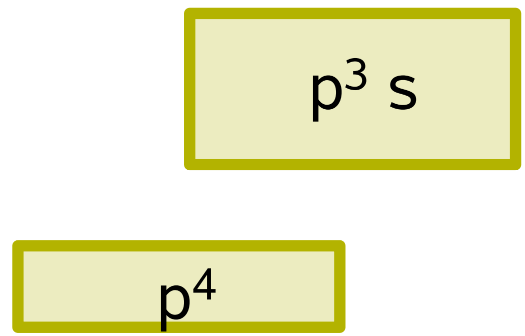
iii) Multiphoton processes in high-Z systems

iv) Nonradiative transitions and autoionization

„Electronic correlations"

-- Fine-structure of open-shell configurations

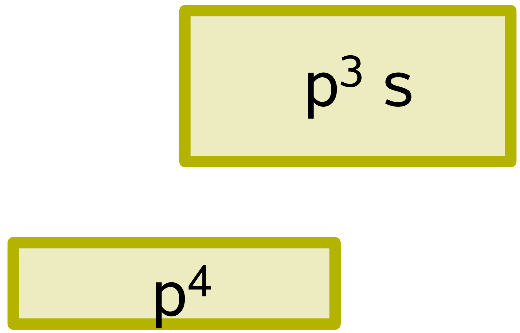
	Dimension of the Hilbert space
p^4	5
$p^3\ s$	10



„Electronic correlations“

-- Fine-structure of open-shell configurations

	Dimension of the Hilbert space
p^4	5
$p^3\,s$	10
d^8	9
$d^7\,p$	110
$f^{\,7}\,s^2$	327
$f^{\,7}\,sp$	3808
$f^{\,7}\,p^2$	4724
$f^{\,6}\,spd$	31804

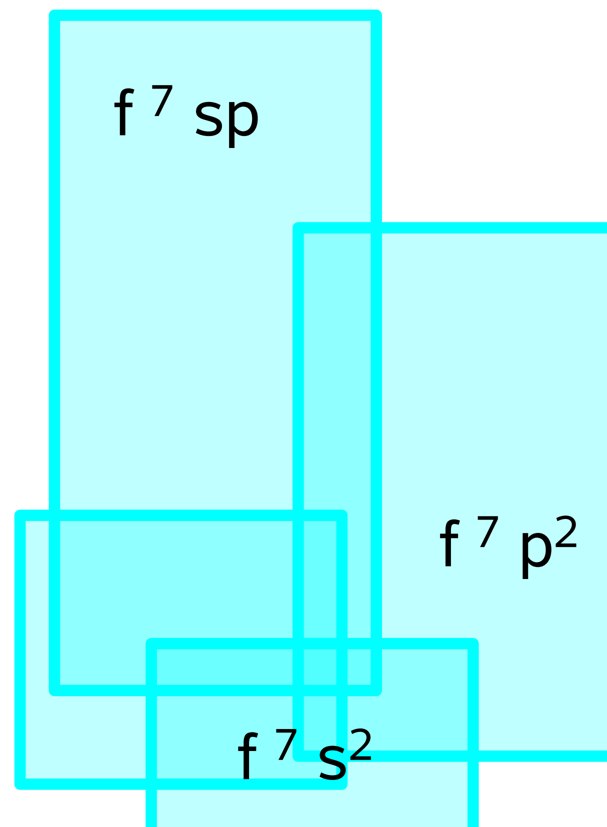


~ 1 .. 3 %

level and transition energies

~ 10 ... 30 %

Computational requirements depend very critically on the shell structure of the atoms and ions !



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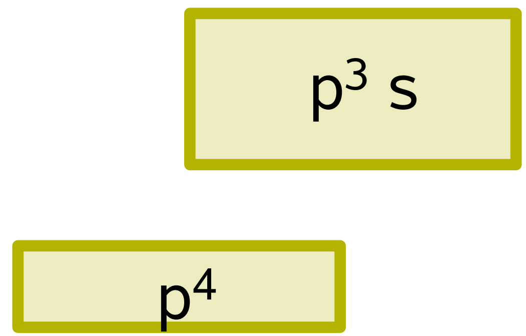

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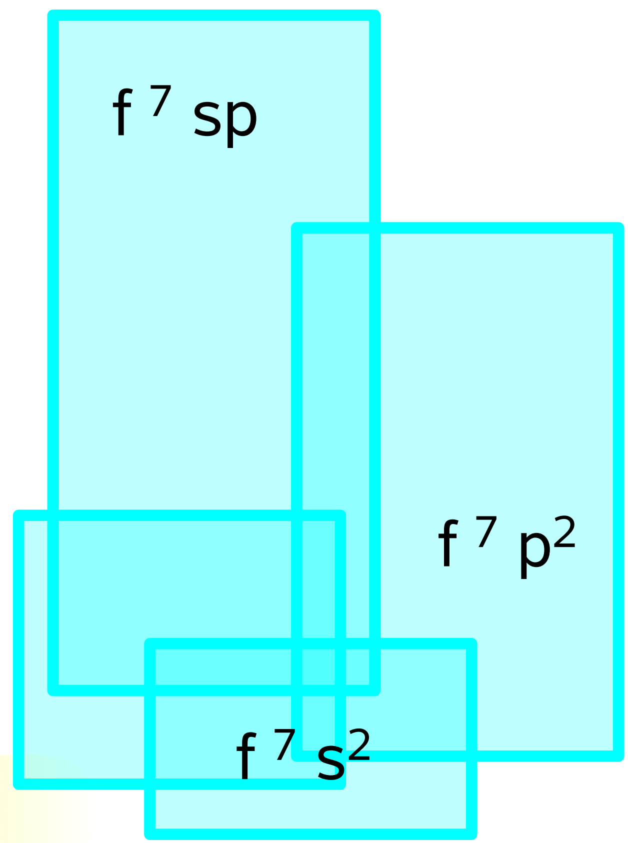
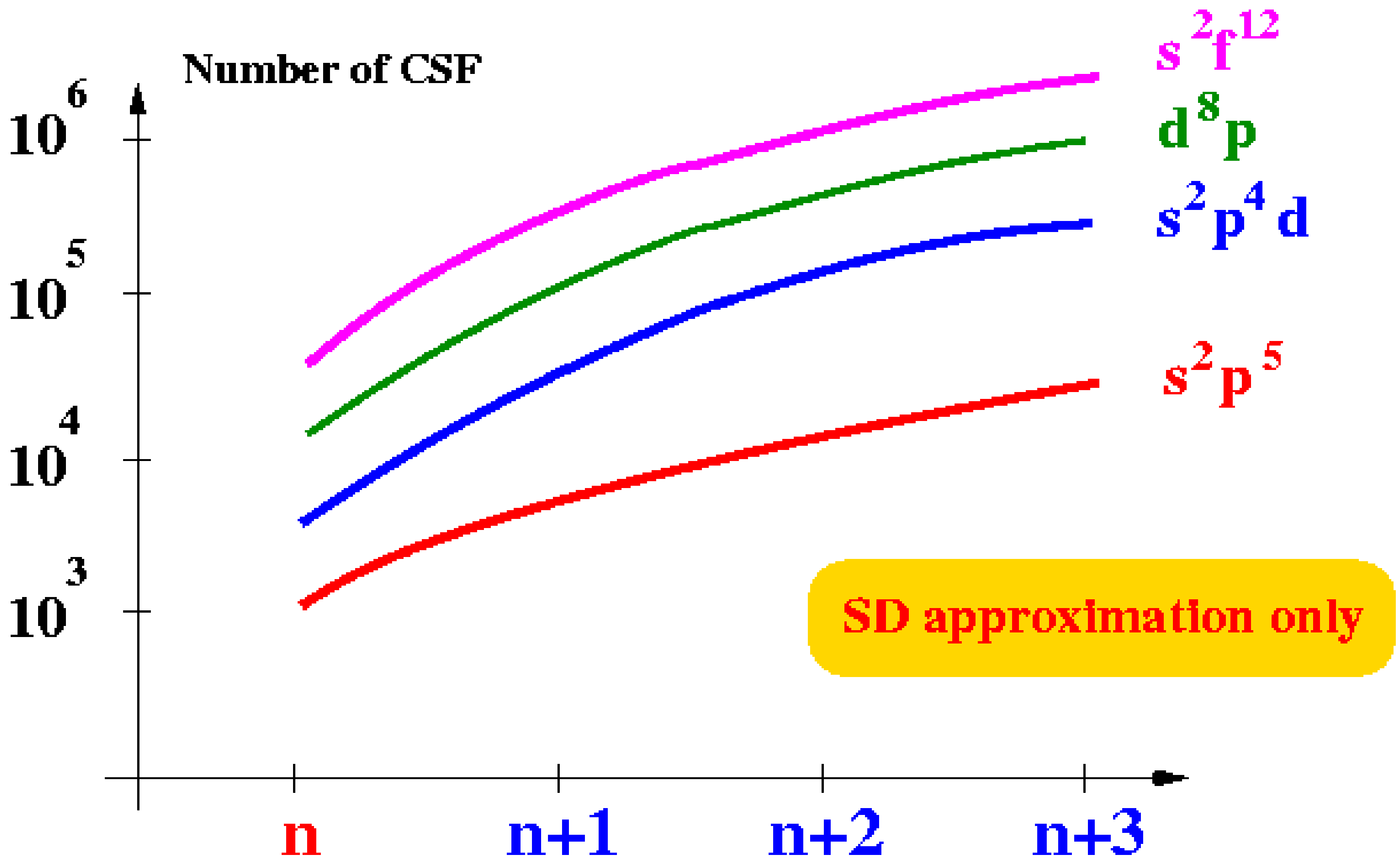
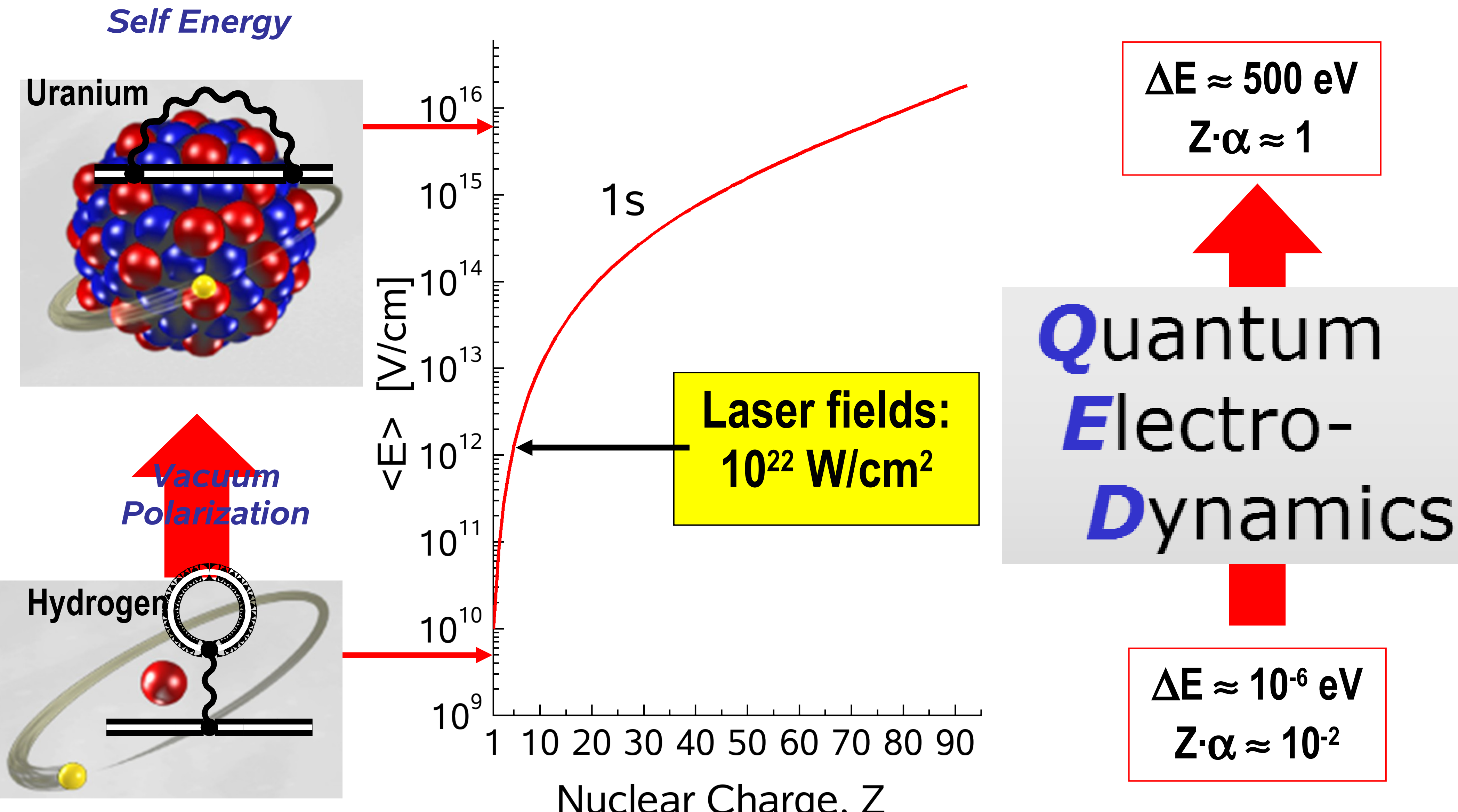


Concept of electron configuration gets lost !

Wave function (CSF) expansions for open-shell structures

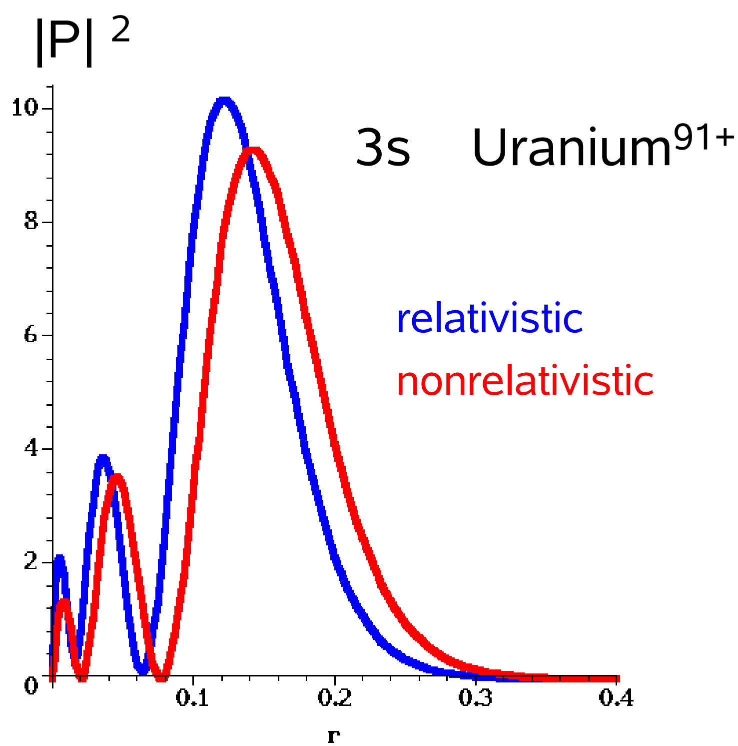


Extreme Static Electromagnetic Fields



Relativistic and quantum-electrodynamical corrections

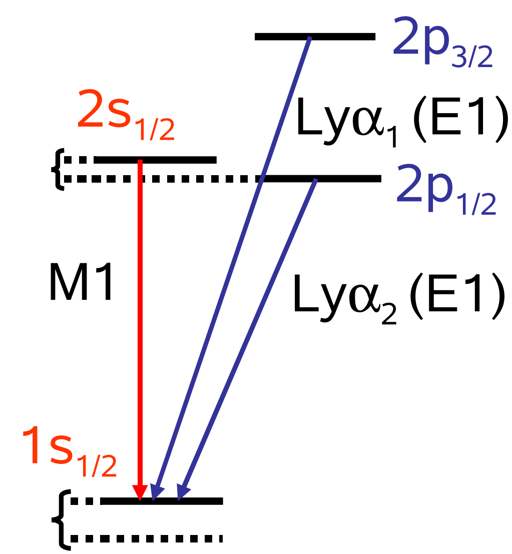
-- Test of QED in hydrogen-like uranium



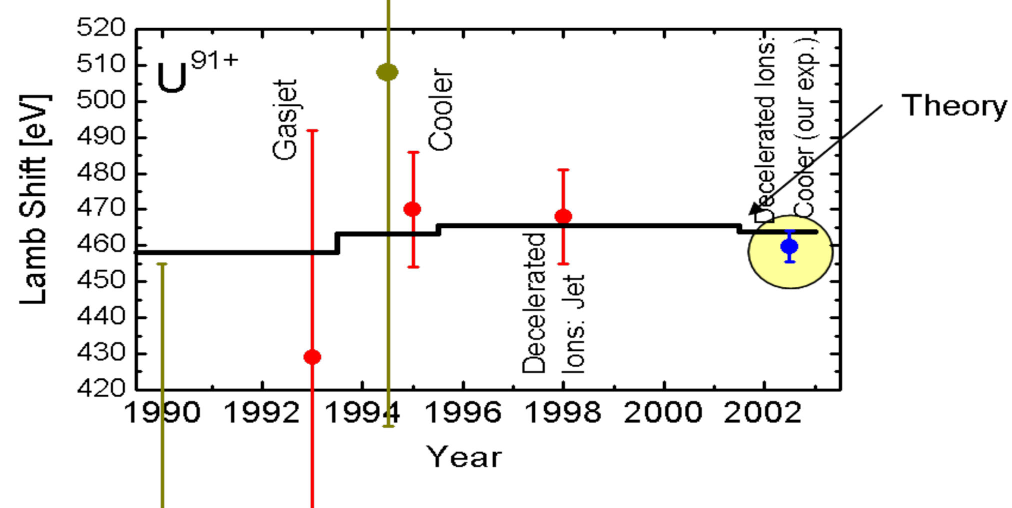
Relativistic contraction
of the wave functions

Use of the one-particle Dirac operator

1s-Lamb Shift	
Experiment: 459.8 eV ± 4.6 eV	
Theory:	463.95 eV



A. Gumberidze, PhD thesis (2003),
PRL 94 (2005) 223001.



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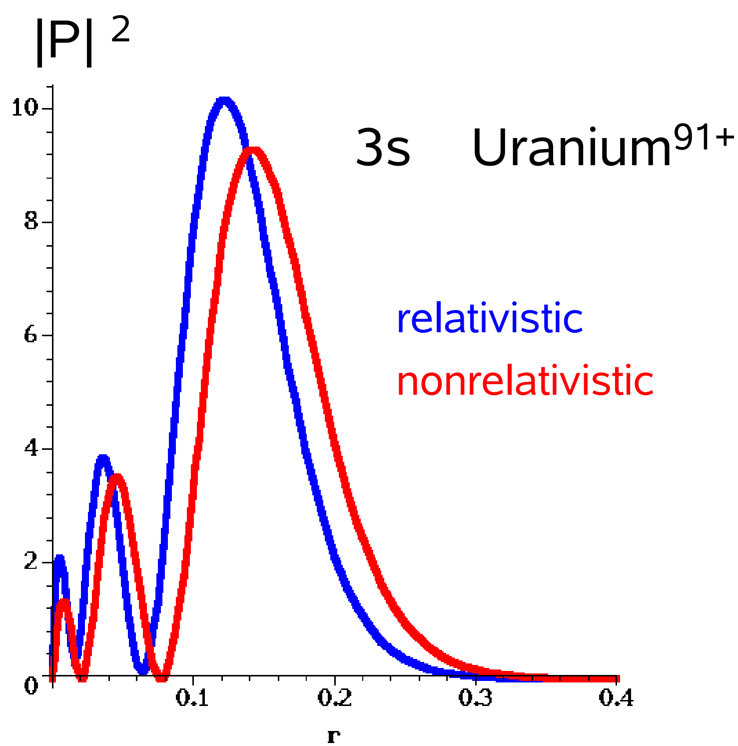

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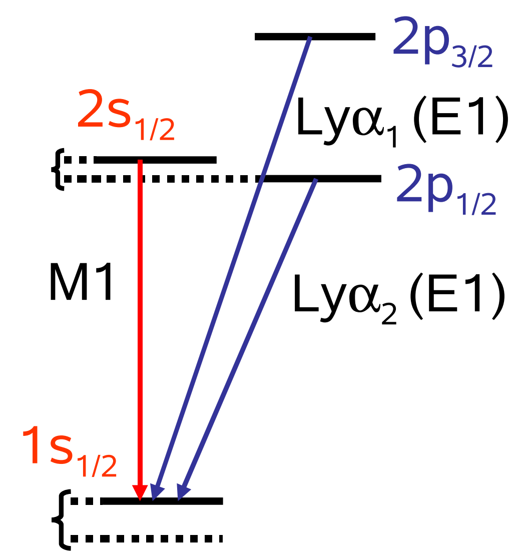

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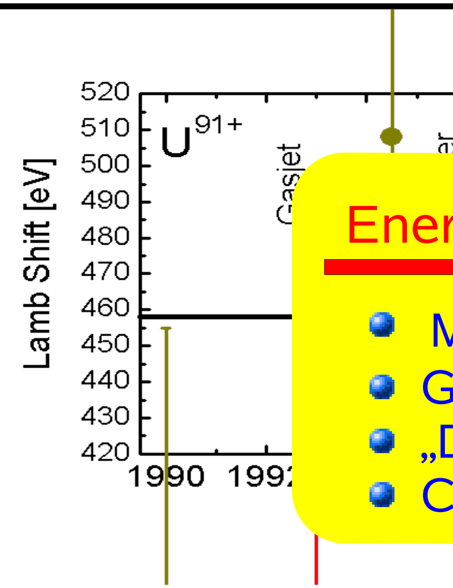


Energies & Wave functions

- MCHF
- GRASP(-92)
- „Desclaux“
- Coupled-Cluster

S. Fritzsche, JESRP 114-116 (2001) 1155; Phys. Scr. T100 (2002) 46

RATIP

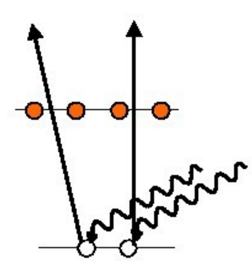
Relativistic Atomic Transition and Ionization Properties

(CPC library)

$$\psi_{\alpha}(P\,J\,M) \;=\; \sum_{r}^{n_c} c_r(\alpha)\,\big|\gamma_r\,P\,J\,M\big\rangle$$

Many-electron basis (wave function expansions)

- Construction and classification of N-particle Hilbert spaces
- Shell model: Systematically enlarged CSF basis
- Interactions
  - Dirac-Coulomb Hamiltonian
  - Breit interactions + QED
  - Electron continuum; scattering phases
- Coherence transfer and Rydberg dynamics



Relativistic CI wave functions including QED estimates and mass polarization

RELCI, CPC 148 (2002) 103

LSJ spectroscopic notation from jj-coupled computations

LSJ, CPC 157 (2003) 239

Auger rates, angular distribu-tions and spin polarization; level widths

AUGER

Photoionization cross sect-ions and (non-dipole) angular parameters

PHOTO

Radiative and dielectronic recombination; angle-angle correlations

...

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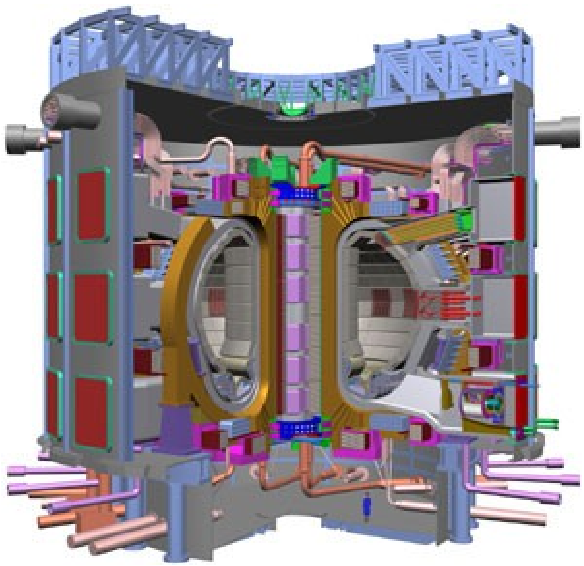


Systematic multiconfiguration calculations

(CI, MCHF, MCDF)

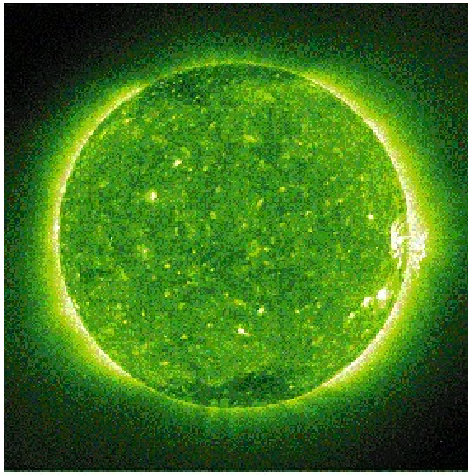
Up to now:

- Term- and hyperfine structure for light elements (Z <=28)
- Resonance and intercombination lines
- Lifetimes

Benchmarks:
He, Li-like, C^{2+}

Example : EUV spectra of multiple-charged iron from the sun

Spectra involving open d-shells



Iron is one of the most abundant heavy elements in the universe(opacity project)

Fe X ... XIV $3s\, 3p^{n+1}$, $3s^2\, 3p^{n-1}\, 3d$

$\Delta E / E < 1\%$

$\Delta A / A = 5 .. 20\ \%$

Improved by factor 5 !

- Line identification
- Improved level structure
- Reliable lifetimes

Fe X: 31 low-lying levels (Dong et al., MNRAS, 1999)
Fe XI: 47 levels (Fritzsche et al., MNRAS, 2000)

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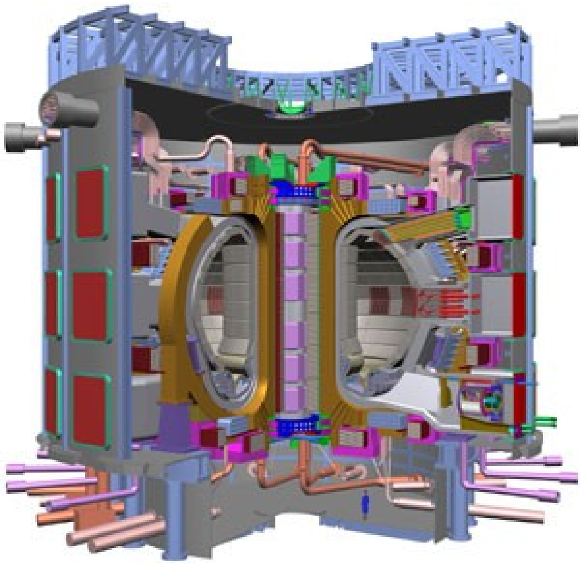
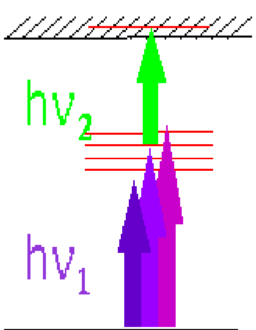
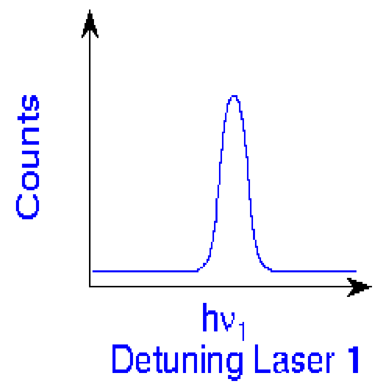


Optical spectroscopy at Fermium (Z = 100)

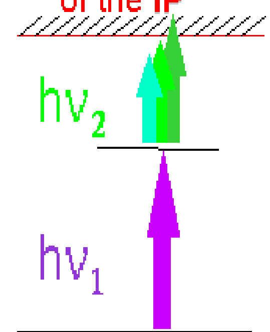
-- first observation and classification of atomic levels

Determination of hfs and isotope shifts





Determination of the IP



Atomic Physics:

- Atomic Structure
- Ionization potentials

Nuclear Physics:

- Nuclear spins
- Moments
- Changes of charge radii

$5f^{12}\,7s^2$, $J^p = 6^+$ $^3H_6^e$

$5f^{12}\,7s\,7p$, $J^p = 6^-, 5$ $^5G_{6,5}^o$

$5f^{12}\,7s\,7p$, $J^p = 6^-, 5^-, 7^-$ $^3H_{6,5,7}^o$ (?)

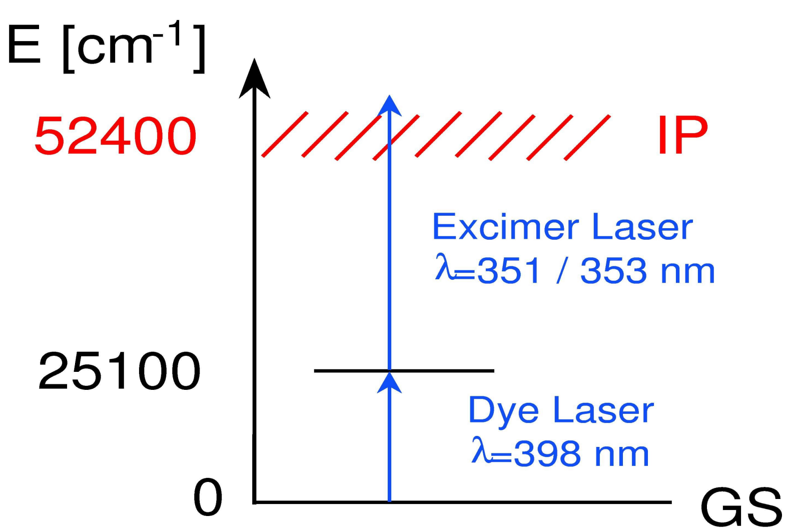


TABLE I. Results of MCDF calculations. Accuracy of transition energy $\overline{\nu}$ is $\Delta\overline{\nu} = \pm 2400\,\text{cm}^{-1}$, A_{ki} = Einstein coefficient, classification according to the largest coefficient c in the CSF expansion.

No.	$\bar{\nu}$ (cm^{-1})	J	A_{ki} s^{-1}	Config.	Term	$\lvert c\rvert^2$
1	0	6	0	$5f^{12}7s^2$	$^3H_6^e$	0.96
2	25 226	6	1.89×10^6	$5f^{12}7s7p$	$^5I_6^o$	0.46
3	25 471	5	1.28×10^6	$5f^{12}7s7p$	$^5G_5^o$	0.34
4	27 394	6	2.43×10^8	$5f^{12}7s7p$	$^3H_6^o$	0.62
5	27 633	5	1.98×10^8	$5f^{12}7s7p$	$^3G_5^o$	0.60
6	27 802	7	3.67×10^8	$5f^{12}7s7p$	$^3I_7^o$	0.66

Zhou & Froese Fischer., Phys. Rev. Lett. 88 (2002) 183001

Low-lying resonances for heavy and super-heavy elements

… for lutetium (Z=71) and lawrencium (Z=103)

TABLE I. The transition energies in cm^{-1} of $nd\ ^2D_{3/2} - (n+1)p\ ^2P^o_{1/2,3/2}$ and the size of CSF expansions for Lu ($n = 5$) and Lr ($n = 6$).

Expansion	$^2D_{3/2} - {}^2P^o_{1/2}$	$^2D_{3/2} - {}^2P^o_{3/2}$	CSF ($^2D_{3/2}/{}^2P^o_{1/2}/{}^2P^o_{3/2}$)
	Lu		
VV + CV($4f^{14}$)	3989	7276	4354/2071/3813
VV + CV($5p^6 4f^{14}$)	8004	11 483	5600/2764/5073
VV + [(CV + CC) ($5p^6 4f^{14}$)]	3857	7130	128 763/36 974/100 277
VV + [(CV + CC) ($4d^{10} 5s^2 5p^6 4f^{14}$)]	4186	7462	305 717/87 241/236 554
RCC [7]	3828	7140	
DFT [10]	3862		
Exp.	4136	7476	
DHF Breit Correction	87	53	
DHF Breit & QED Correction	76	43	
	Lr		
VV + CV($5f^{14}$)	−1298	9137	3659/1842/3338
VV + CV($6p^6 5f^{14}$)	1339	12 761	4708/2495/4495
VV + [(CV + CC) ($6p^6 5f^{14}$)]	−1953	6469	125 325/37 333/97 500
VV + [(CV + CC) ($5d^{10} 6s^2 6p^6 5f^{14}$)]	−1127	7807	330 252/95 969/246 376
RCC	−1388	6960	
RCC with Breit	−1263	7010	
DHF Breit Correction	97	4	
DHF Breit & QED Correction	59	−26	

RCC: Eliav et al.., Phys. Rev. A52 (1995) 291; DFT: Vosko & Chevary, J. Phys. B26 (1993) 873

Low-lying resonances for heavy and super-heavy elements

-- oscillator strengths in different gauges

TABLE II. The oscillator strengths of $nd\ ^2D_{3/2} - (n+1)p\ ^2P^o_{1/2,3/2}$ for Lu ($n = 5$) and Lr ($n = 6$).

Expansion	$^2D_{3/2} - {}^2P^o_{1/2}$ gf_L	gf_V	Scaled gf_L	$^2D_{3/2} - {}^2P^o_{3/2}$ gf_L	gf_V	Scaled gf_L
			Lu			
VV + CV($4f^{14}$)	0.0304	0.0582	0.0315	0.0111	0.0219	0.0114
VV + CV($5p^6 4f^{14}$)	0.0511	0.1552	0.0264	0.0144	0.0467	0.0094
VV + [(CV + CC)($5p^6 4f^{14}$)]	0.0908	0.3835	0.0974	0.0322	0.0856	0.0337
VV + [(CV + CC)($4d^{10}5s^2 5p^6 4f^{14}$)]	0.1043	0.3345	0.1031	0.0354	0.0742	0.0355
			Lr			
VV + CV($5f^{14}$)	−0.0162	−0.0076		0.0210	0.0313	
VV + CV($6p^6 5f^{14}$)	0.0144	0.2359		0.0227	0.0839	
VV + [(CV + CC)($6p^6 5f^{14}$)]	−0.0624	−0.0002		0.0414	0.0867	
VV + [(CV + CC)($5d^{10}6s^2 6p^6 5f^{14}$)]	−0.0378	−0.0024		0.0519	0.0685	

Good accuracy of the (atomic) energies is a necessary, but not a sufficient criterion !

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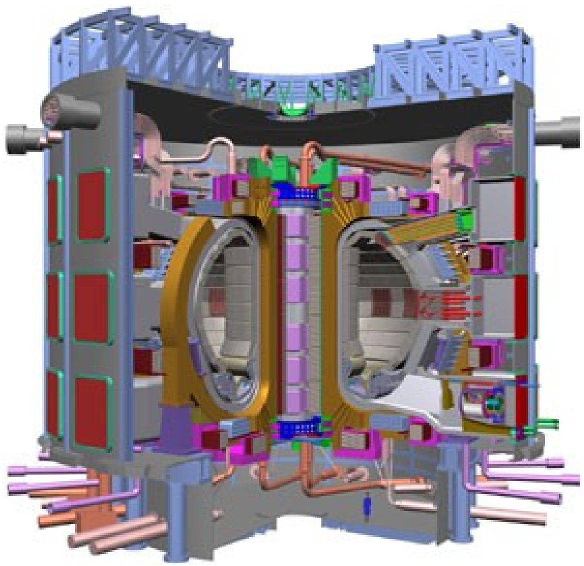
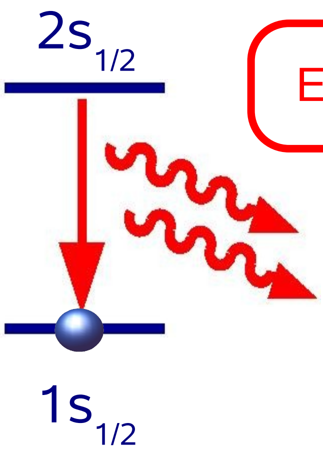
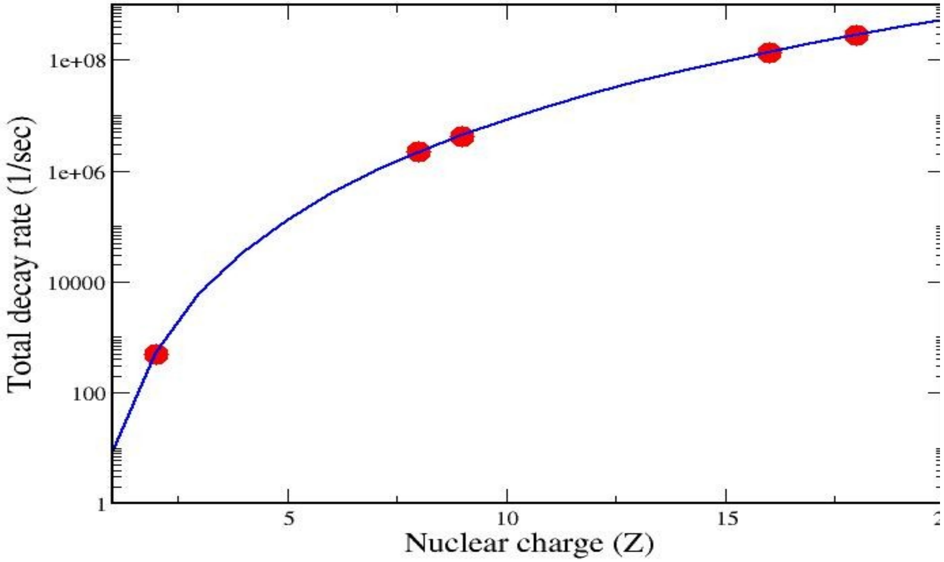


Two-photon decay of highly-charged ions



E1E1

$$\Gamma_{tot} \approx \Gamma_{E1E1} = 8.229 \cdot Z^6$$



A.Surzhykov et al. PRA 71 (2004) 012034

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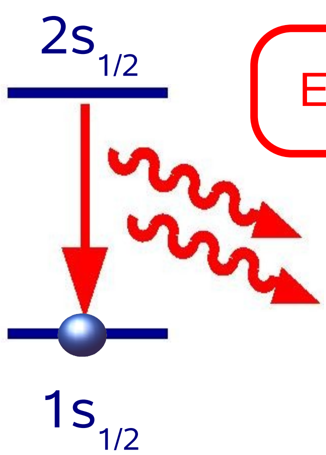
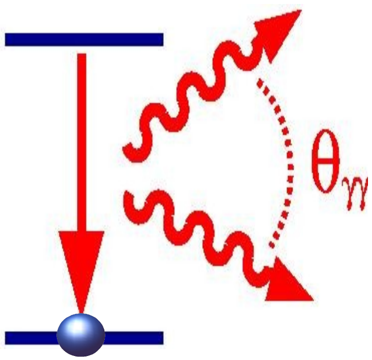
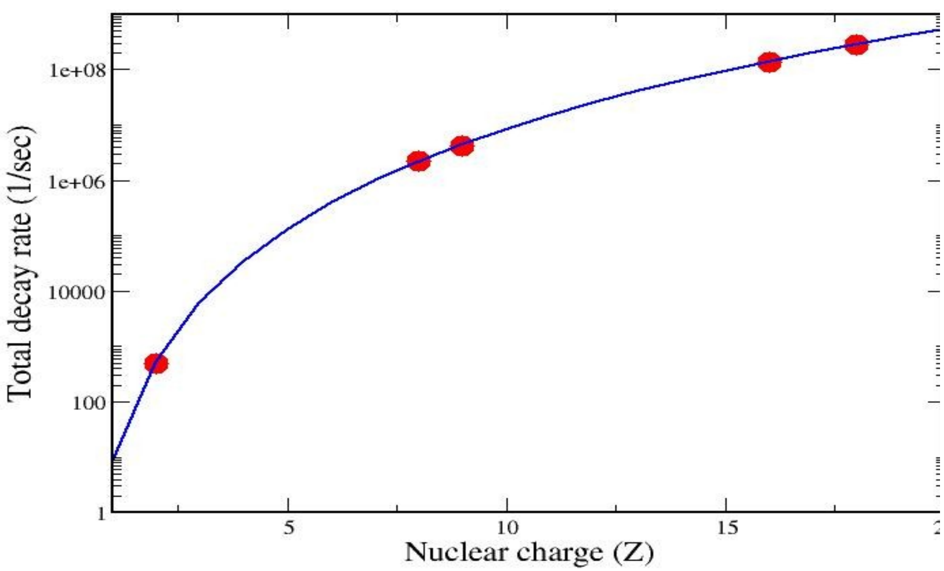


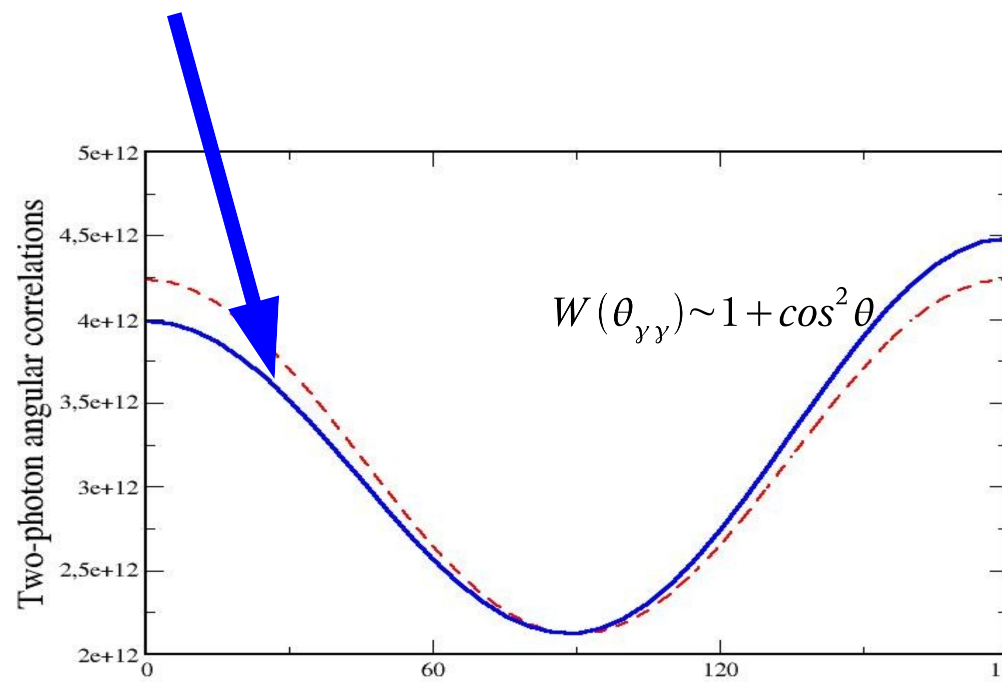
E1E1 + E1M2 + M1M1+E2E2 + E2M1....

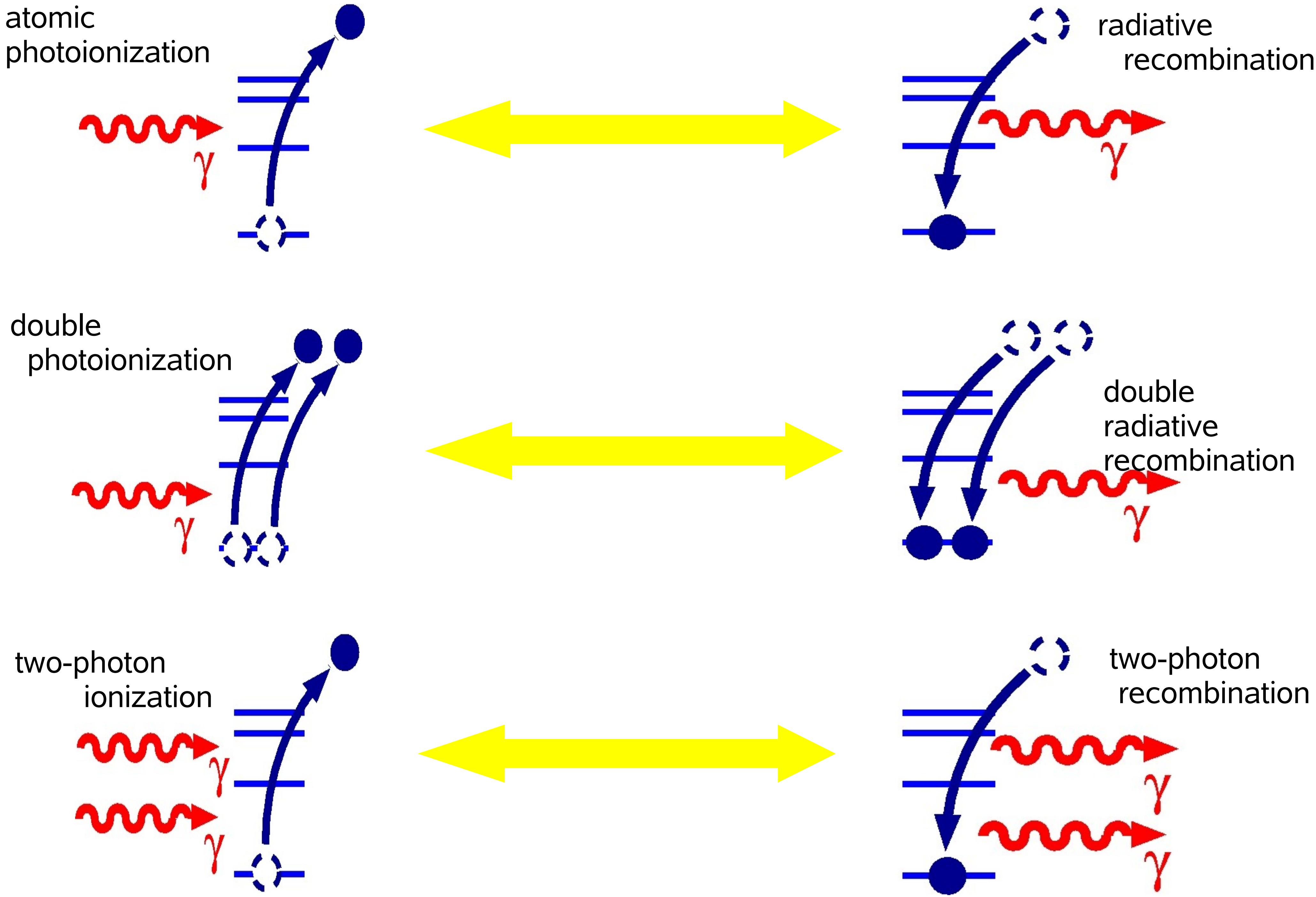


Higher multipoles give rise to an asymetrical shift

$$\Gamma_{tot} \approx \Gamma_{E1E1} = 8.229 \cdot Z^6$$






atomic photoionization
γ
radiative recombination
γ
double photoionization
γ
double radiative recombination
γ
two-photon ionization
γ
γ
two-photon recombination
γ
γ

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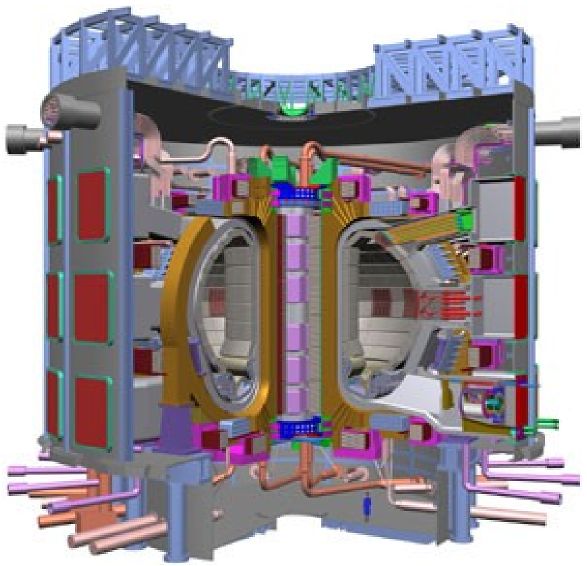
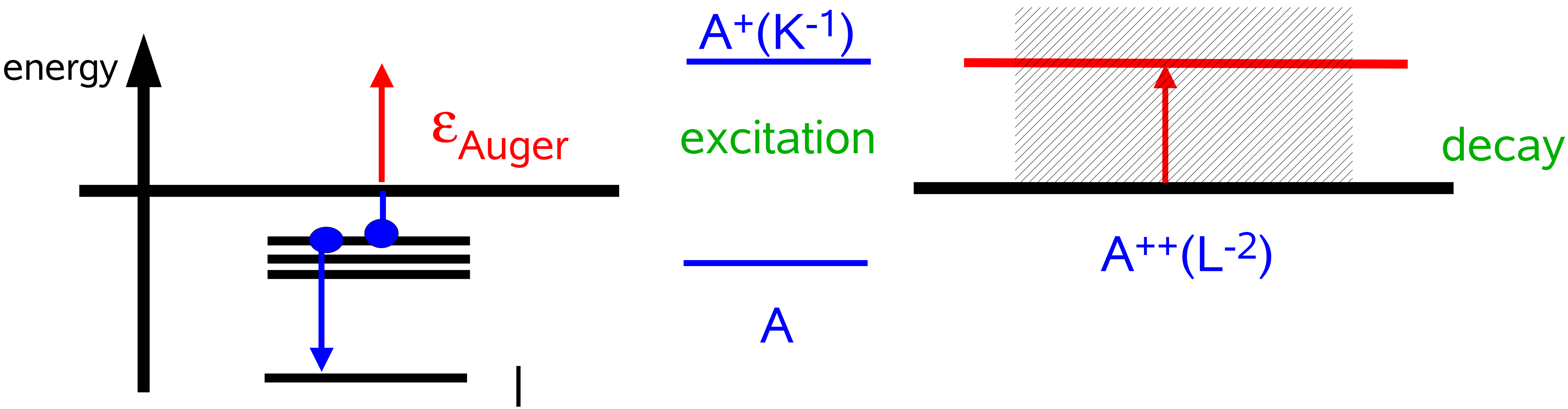


Auger emission of excited atomic states



$$H = \sum_i \left(h_i + u(r_i)\right) \qquad H = \sum_i h_i + \sum_{i<j} \frac{1}{r_{ij}}$$

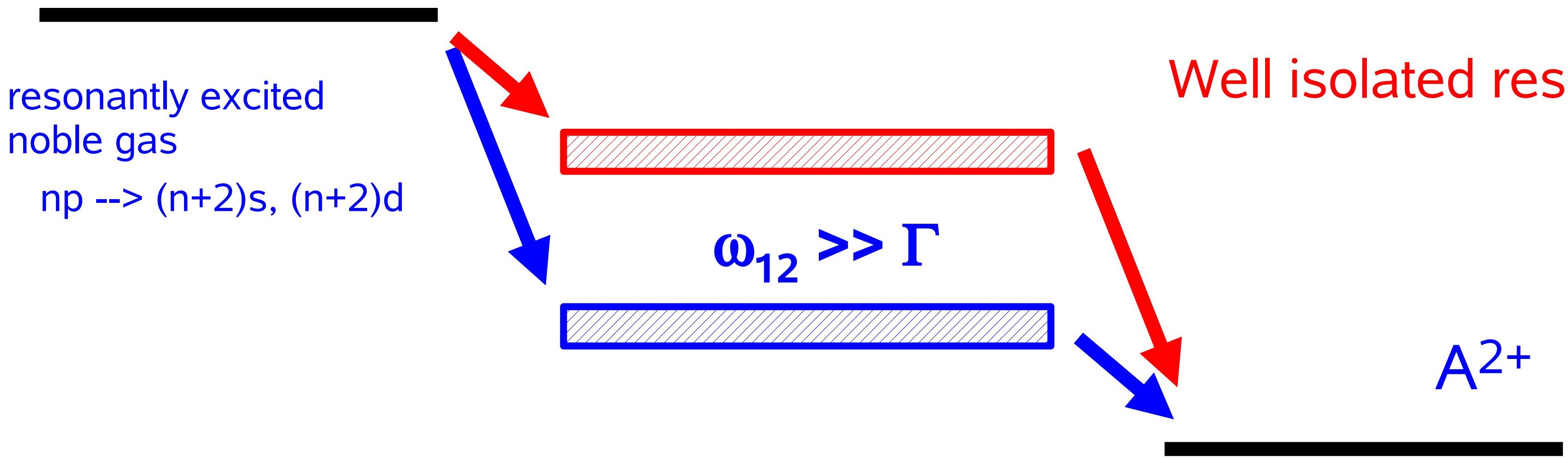
Wentzel's ansatz: Autoionization is caused by electron-electron interactions which cannot be considered in an one-particle picture.

$$\sum_{i<j} \frac{1}{r_{ij}} - \sum_i u(r_i)$$

Ideal tool for a better understanding of electronic correlations !

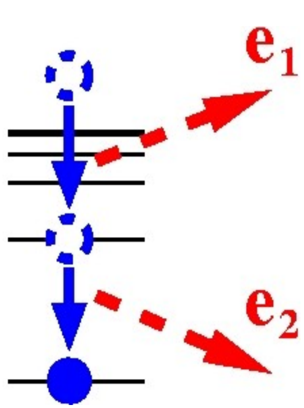
Coherence transfer in the Auger cascades of noble gases

-- a signature of the „atomic double slit“



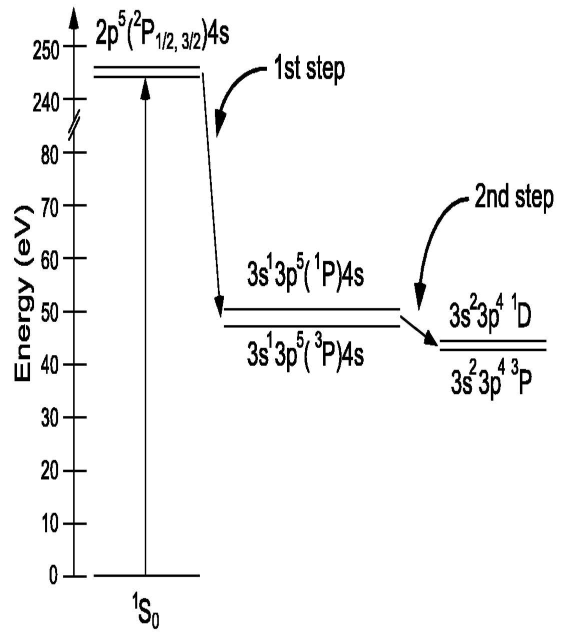
Well isolated resonances !

Decay branches are independent; „path“ can be determined by measuring the energy spectrum.



Collaboration with Nicolai Kabachnik (Bielefeld);
experiments by Kyioshi Ueda and coworkers at SPring8, Japan

Excitation and two-step Auger cascades in noble gases



Photoabsorption:

$$\mathrm{Ar}\ (2p^6\ 3s^2\ 3p^6\ ^1S_0) + h\nu \longrightarrow \mathrm{Ar}^*(2p^5\ 3s^2\ 3p^6\ 4s\ ^1P_1)$$

First decay:

$$\mathrm{Ar}^*(^1P_1) \longrightarrow \mathrm{Ar}^{*+}(3s\ 3p^5\ (^{1,3}P)\ 4s\ ^2P \text{ or } ^4P) + e_{A1}$$

Second decay:

$$\mathrm{Ar}^{*+}\ (3s\ 3p^5\ (^1P)\ 4s\ ^2P_{1/2,3/2}) \longrightarrow \mathrm{Ar}^2+\ (3p^4\ ^3P \text{ or } ^1D) + e_{A2}$$

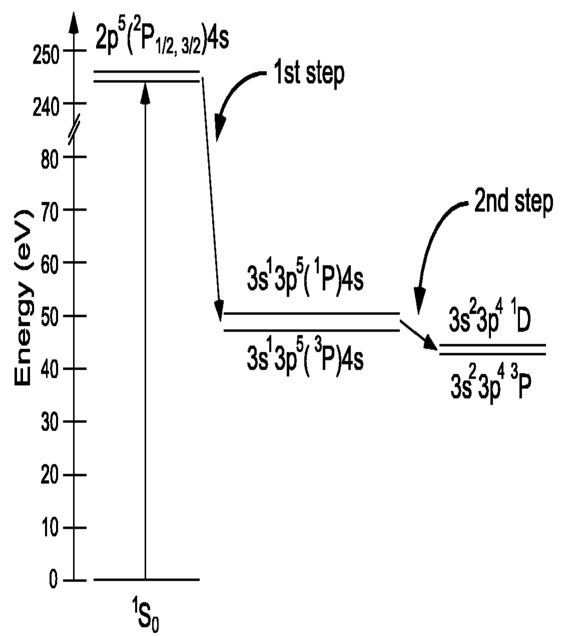
Ne:	500 : 1
Ar:	80 : 1
Kr:	25 : 1
Xe:	8 : 1

$$\frac{A_{resonance}}{A_{intercombination}}$$

Xenon: $4d^{-1}6p\ ^{1,3}P_1 \longrightarrow 5s^{-2}6p;\ 5s5p^56p$

Relativity enters here in two ways !

Excitation and two-step Auger cascades in noble gases



Photoabsorption:

$Ar\ (2p^6\ 3s^2\ 3p^6\ ^1S_0) + h\nu \rightarrow Ar^*(2p^5\ 3s^2\ 3p^6\ 4s\ ^1P_1)$

First decay:

$Ar^*(^1P_1) \rightarrow Ar^{*+}(3s\ 3p^5\ (^{1,3}P)\ 4s\ ^2P \text{ or } ^4P) + e_{A1}$

Second decay:

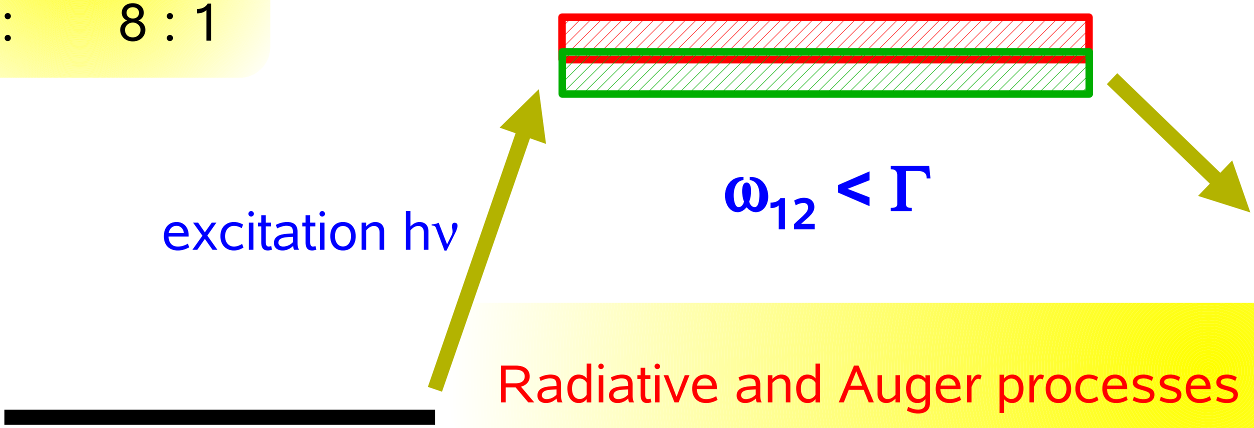
$Ar^{*+}\ (3s\ 3p^5\ (^1P)\ 4s\ ^2P_{1/2,3/2}) \rightarrow Ar^{2}+\ (3p^4\ ^3P \text{ or } ^1D) + e_{A2}$

Ne:	500 : 1
Ar:	80 : 1
Kr:	25 : 1
Xe:	8 : 1

$$\frac{A_{resonance}}{A_{intercombination}}$$

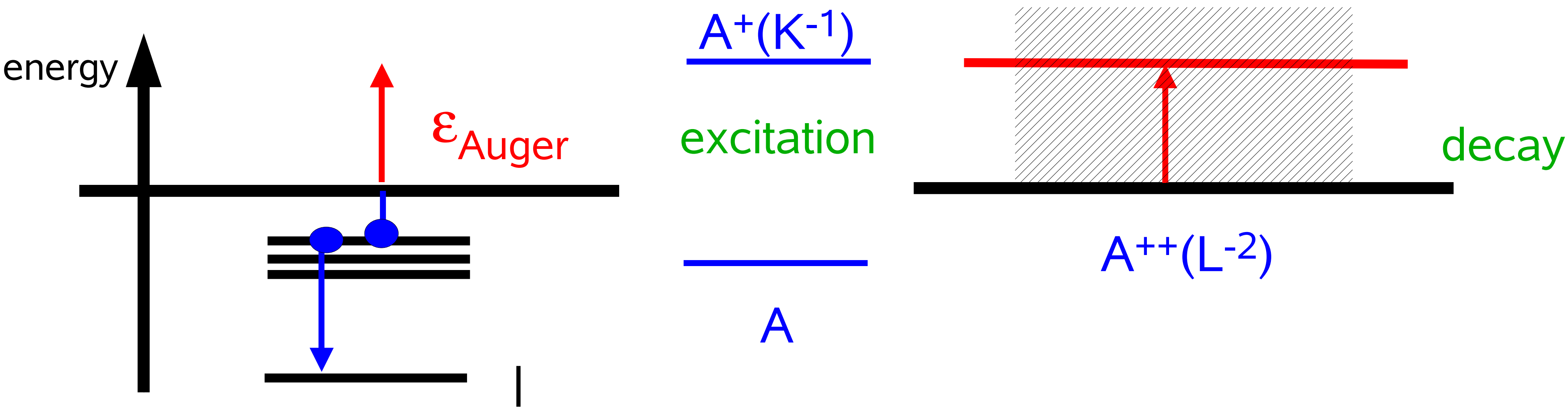
Xenon: $4d^{-1}6p\ ^{1,3}P_1 \rightarrow 5s^{-2}6p;\ 5s5p^56p$

Kitajima et al., JPB 34 (2001) 3829; JPB 35 (2002) 3327.



Radiative and Auger processes are not longer independent !

Auger emission of excited atomic states



$$H_{DCB} = \sum_i h_D(i) \; + \; \sum_{i<j} \frac{1}{r_{ij}} \; + \; \sum_{i<j} \frac{1}{2r_{ij}} \left[\alpha_i \alpha_j \; + \; \frac{(\alpha_i r_i)(\alpha_j r_j)}{r_{ij}^2} \right]$$

Wentzel's ansatz: Autoionization is caused by electron-electron interactions which cannot be considered in an one-particle picture.

$$\sum_{i<j} \frac{1}{r_{ij}} + b(i,j) \; - \; \sum_i u(r_i)$$

Breit interaction

Summary and outlook

- Accurate atomic data are needed (more or less urgently) for a wide range of applications.
- Atomic physics still provides a great „playground“ for studying many-particle processes and electronic correlations.
- New numerical techniques have to meet the requirements for a whole „class of systems“ and not only provide 'proofs of principle'.
- Complexity of (atomic) many-particle systems: Development of ab-initio methods cannot always be separated from the processes and properties; overlap with experimental progress.
- Present and future challenges:

  Improved treatment of open-shell structures and highly excited states

  Coupling of bound-state densities to the continuum

  (capture and emission of electrons, multi-photon processes, Fano resonances, „complete experiments“)