Correlated quantum systems in intense fields

– dynamics of multiple and highly-charged ions

S. Fritzsche

6th February 2013

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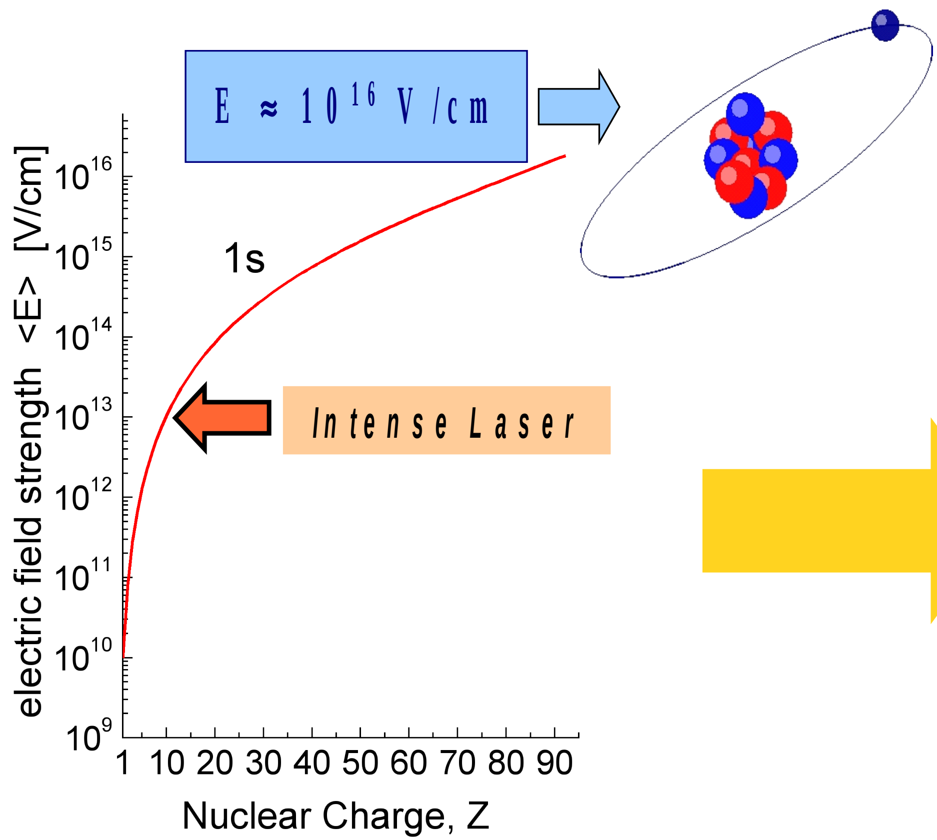
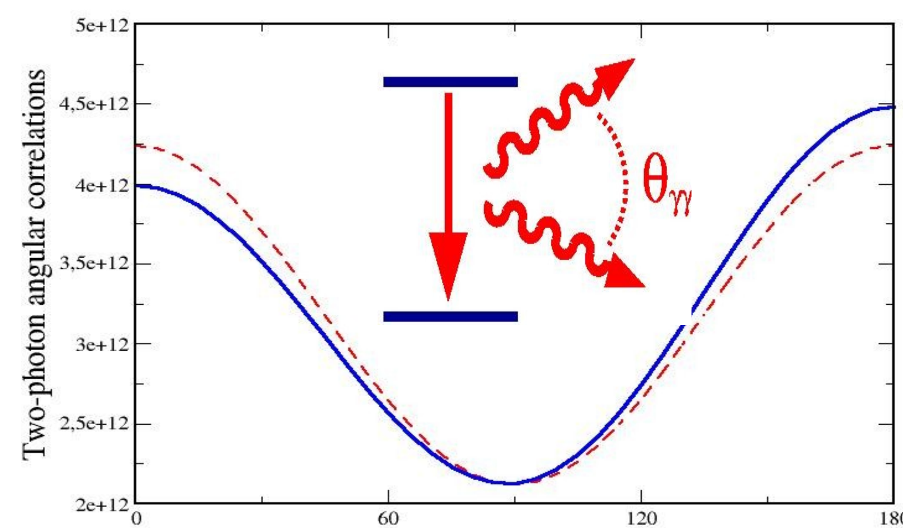
- Ionization & recombination in strong Coulomb fields

correlated photon and/or electron emission



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



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

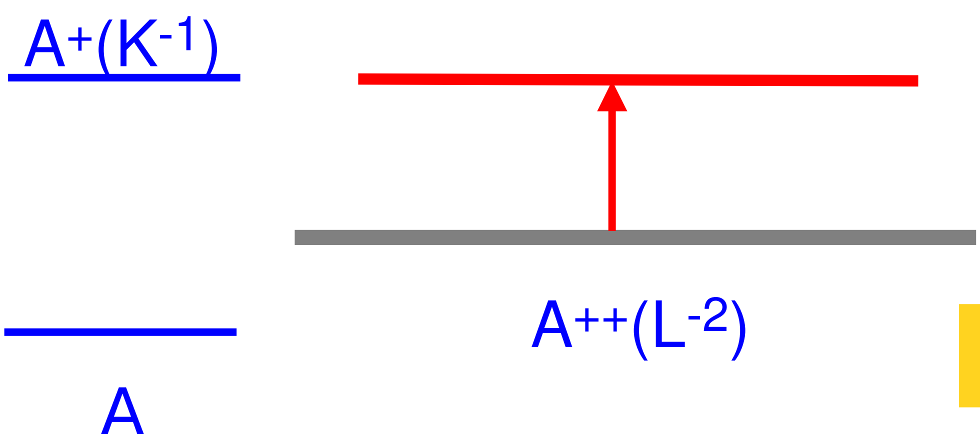
Correlated quantum systems in intense fields

- Ionization & recombination in strong Coulomb fields

  correlated photon and/or electron emission

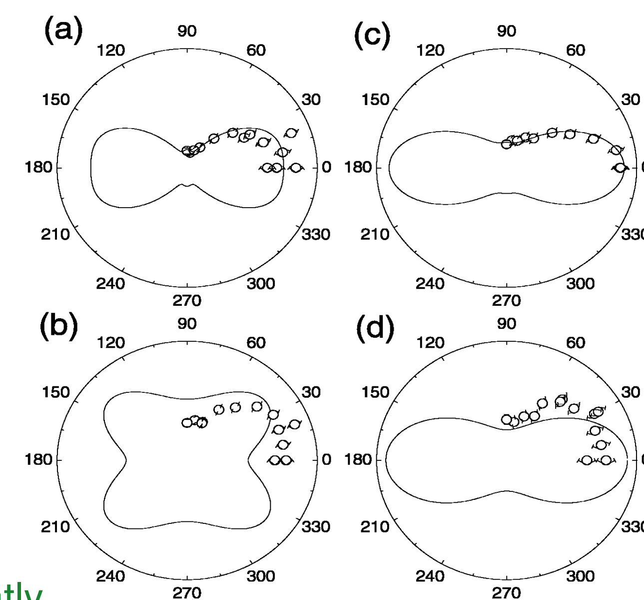
- Coupling of deeply-bound electrons to the continuum

  correlation-induced autoionization & capture



$$H = \sum_i h_i + \sum_{i<j} \frac{1}{r_{ij}}$$

Angular correlations between the subsequently emitted Auger electrons.

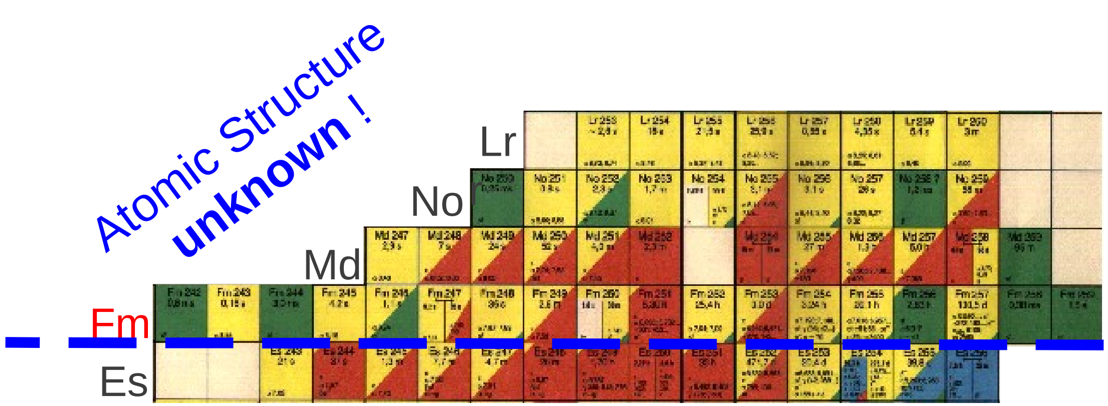


Ueda et al., JPB 36 (2003) 319.

Correlated quantum systems in intense fields

- ...
- Properties of many-electron & (super-) heavy elements

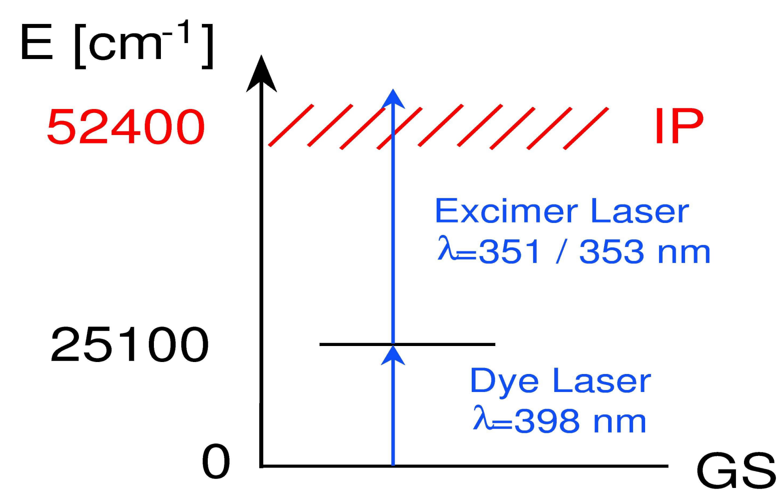
correlated (ab-initio) many-body techniques



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

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

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



M. Sewtz et al., Phys. Rev. Lett. 90 (2003) 163002

Correlated quantum systems in intense fields

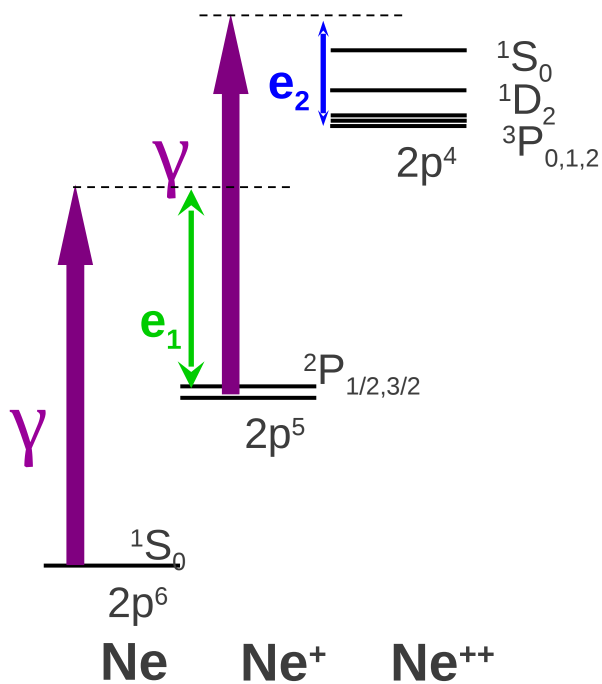
– dynamics of multiple and highly-charged ions

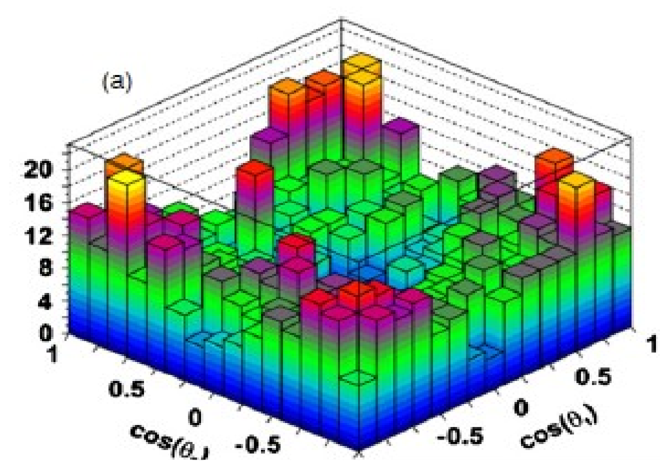
- Properties of many-electron & (super-) heavy elements

  correlated (ab-initio) many-body techniques

- Multi-photon & electron dynamics in intense FEL radiation

  correlated many-electron dynamics





$W(\theta_1, \phi_1=0; \theta_2, \phi_2=0)$

M. Kurka et al, J. Phys. B 42 (2009) 141002.

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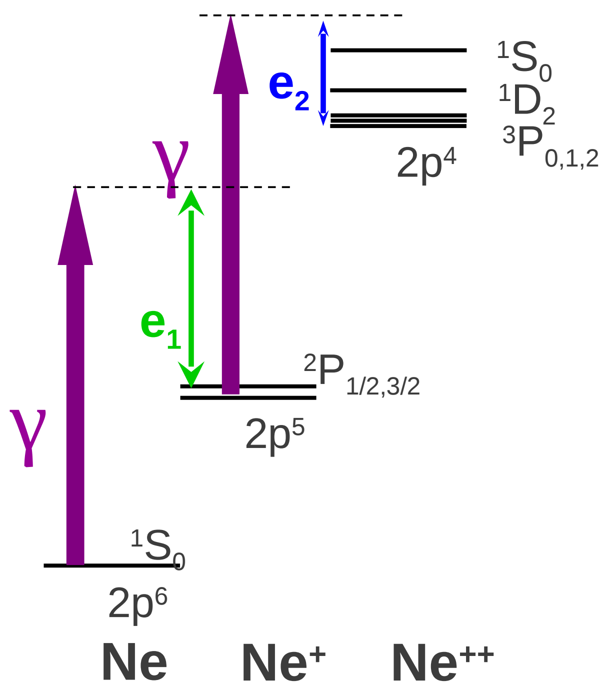

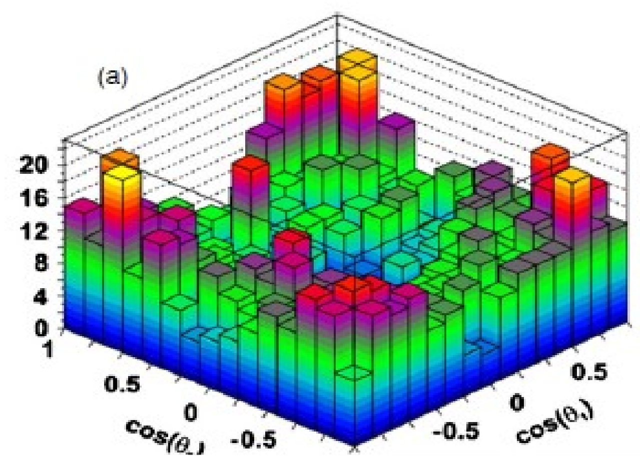

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M. Kurka et al, J. Phys. B 42 (2009) 141002.

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  correlated photon and/or electron emission

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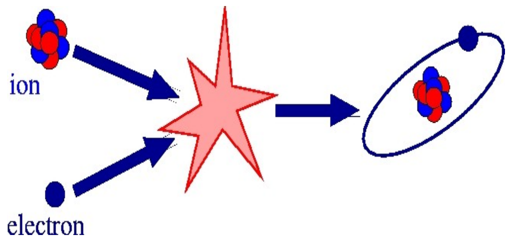
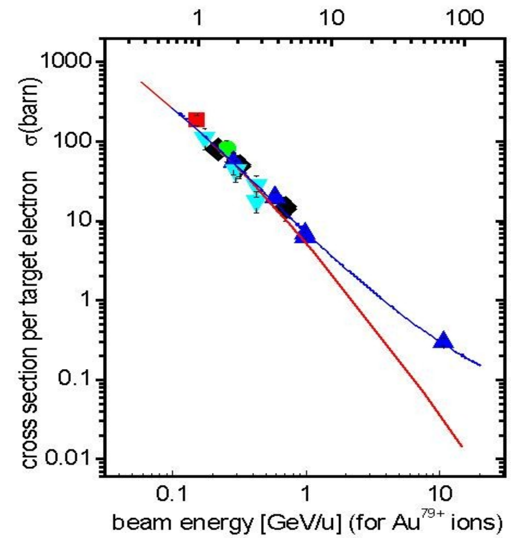
Ionization & recombination in strong Coulomb fields

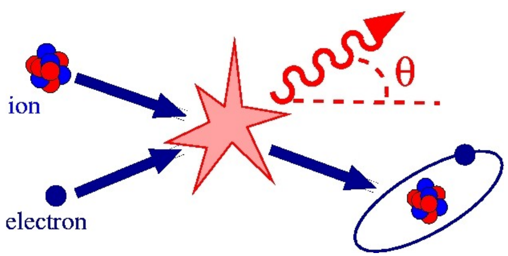
– exploring the light-matter interactions



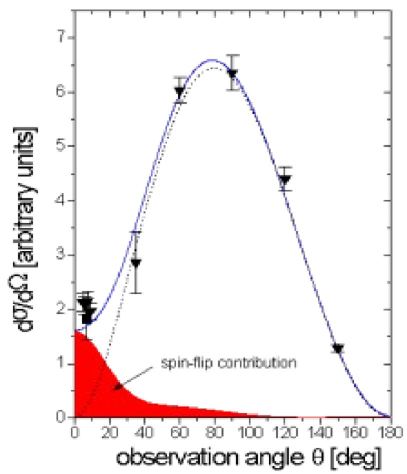
total cross sections

$$\sigma \sim \sum_{polarization} \int d\Omega \, |M|^2$$





angular distributions



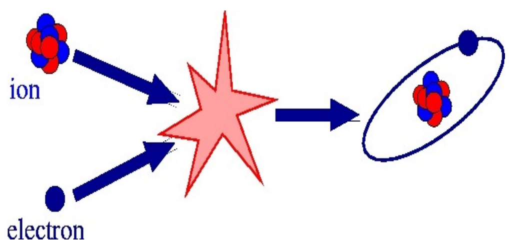
$$\frac{d\sigma}{d\Omega}(\theta) \sim \sum_{polarization} |M|^2$$

photoionization

recombination

Ionization & recombination in strong Coulomb fields

– exploring the light-matter interactions



total cross sections

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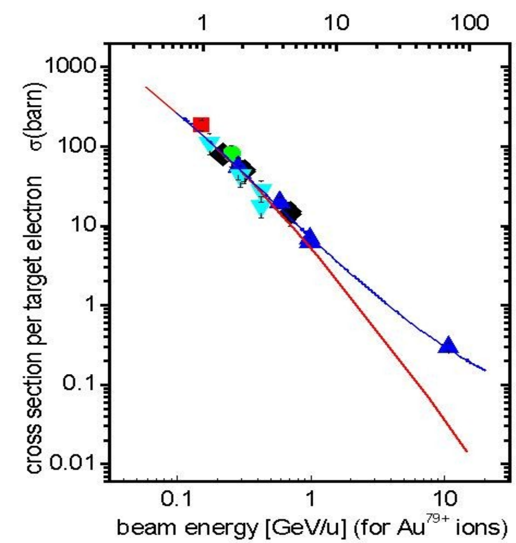
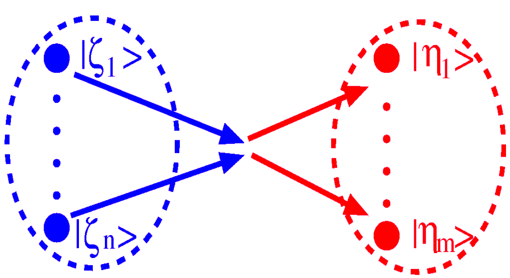


Initial state

$(t \to -\infty)$

$\hat{\rho}_i$



Final state

$(t \to +\infty)$

$\hat{\rho}_f$

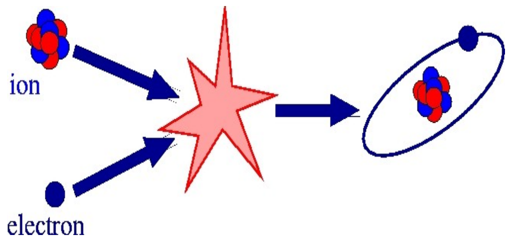
$\hat{S}$ - scattering operator

$$\boxed{\hat{\rho}_f = \hat{S}\,\hat{\rho}_i\,\hat{S}^+}$$

ρ = (μ_S, J, J'; E; I, μ_I ... density matrix)

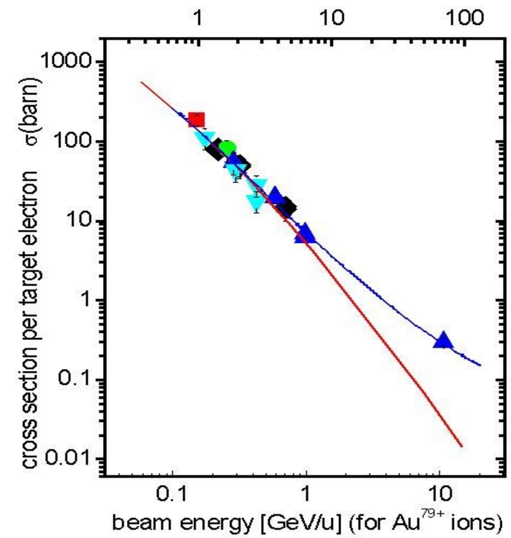
Ionization & recombination in strong Coulomb fields

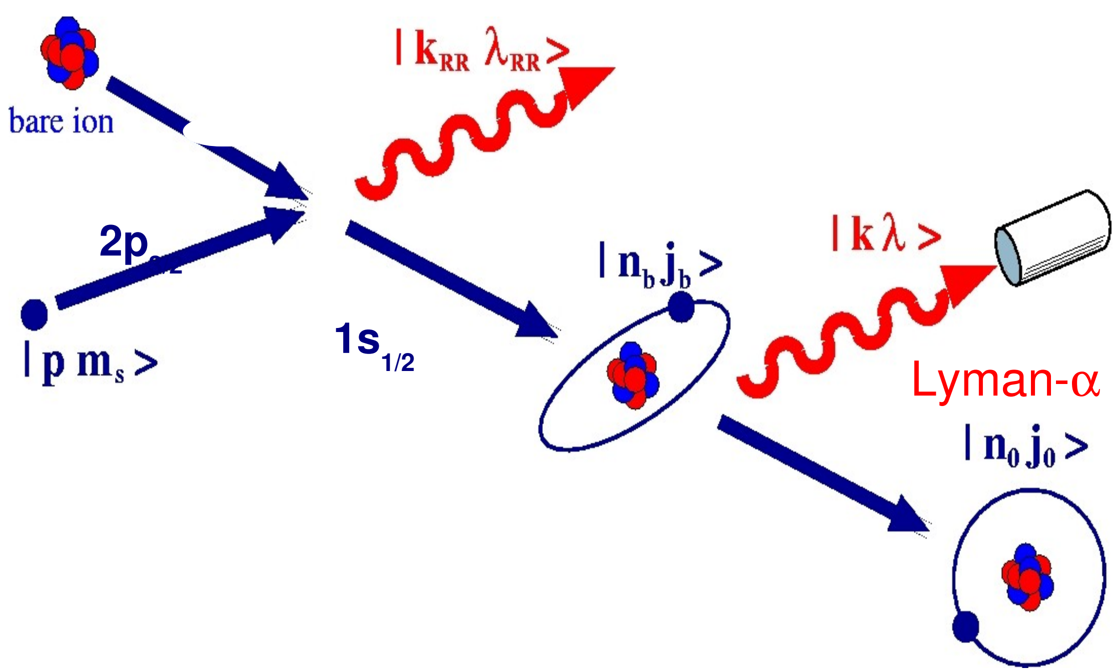
– exploring the light-matter interactions

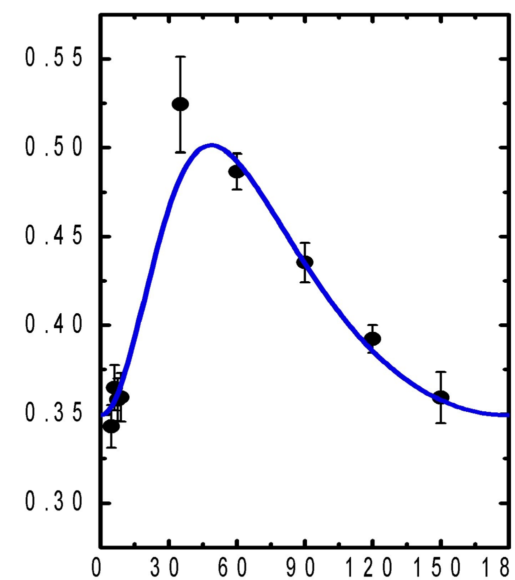


total cross sections

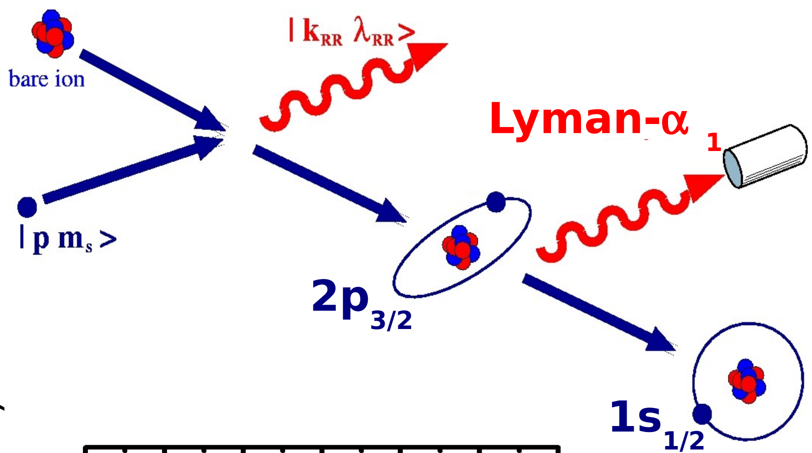
$$\sigma \sim \sum_{polarization} \int d\,\Omega \left| M \right|^2$$



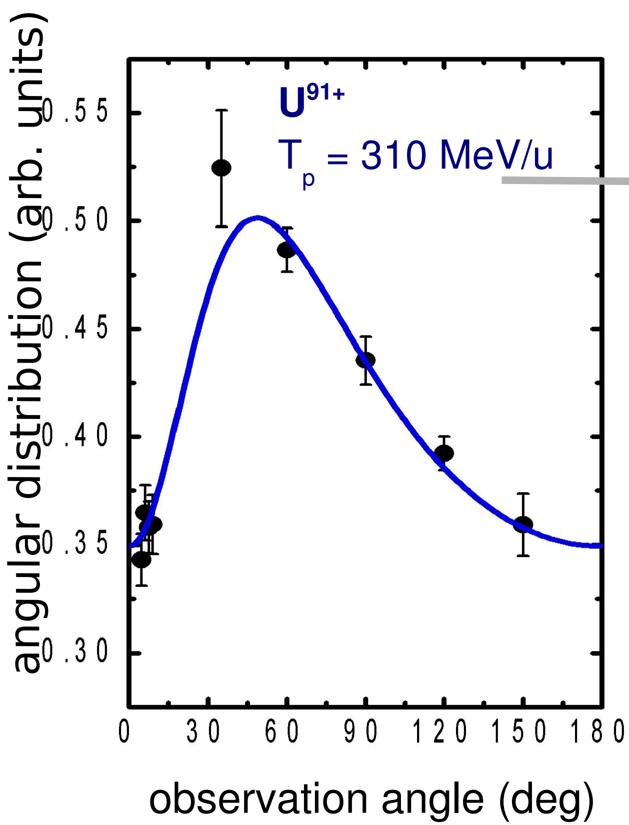




Capture into the $2p_{3/2}$ excited states of initially bare ions

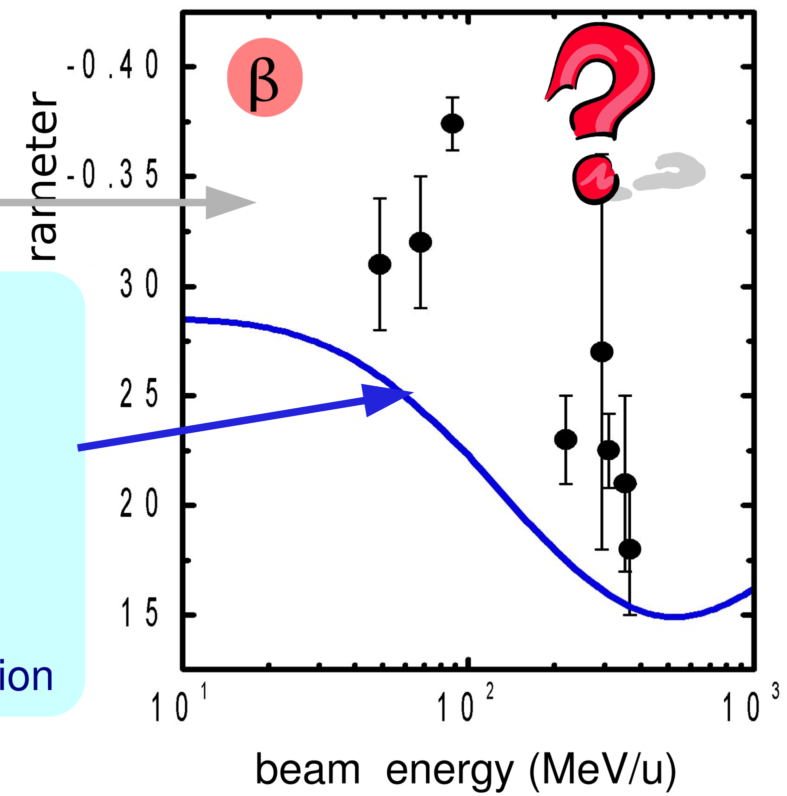


- Magnetic sublevel population of the residual ion can not be measured **directly**
- **But:** knowledge on population of excited ion state may be derived from the properties of subsequent decay



fitting

$$W(\theta) \propto 1 + \beta P_2(\cos\theta)$$



Theory:

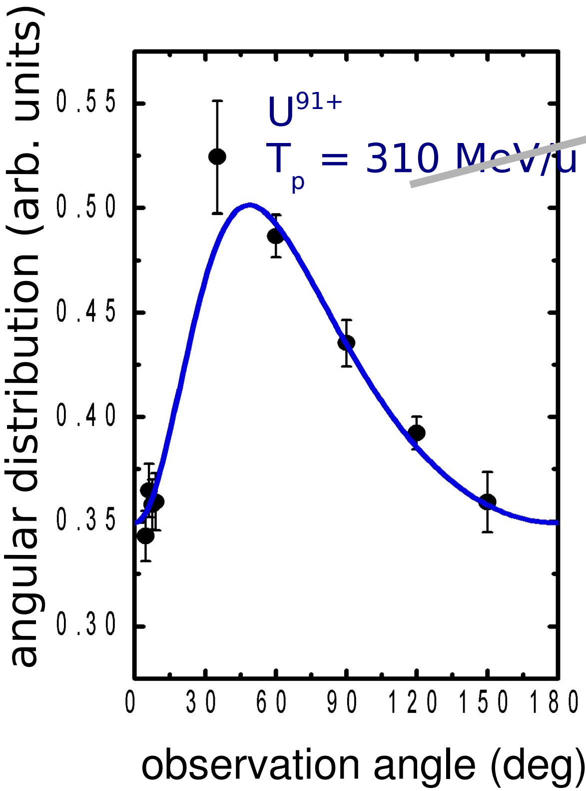
$$\beta = \frac{1}{2}\underbrace{\frac{\sigma_{\mu_b=\pm 3/2} - \sigma_{\mu_b=\pm 1/2}}{\sigma_{\mu_b=\pm 3/2} + \sigma_{\mu_b=\pm 1/2}}}$$

alignment of the $2p_{3/2}$ state:
relative sublevel | j_b m_b> population

Th. Stöhlker et al., PRL 79 (1997) 3270.

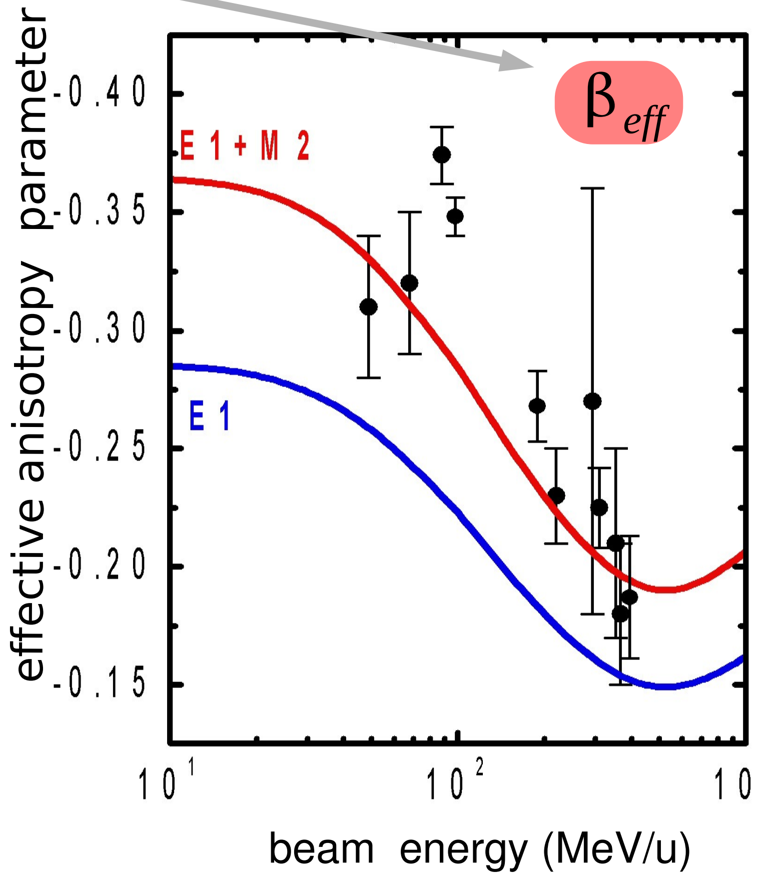
J. Eichler et al., PRA 58 (1998) 2128.

E1-M2 multipole mixing: Alignment of the $2p_{3/2}$ state



fitti
ng

$$W(\theta) \propto 1 + \beta_{eff} P_2(\cos\theta)$$



Dynamical alignment studies enables one to explore magnetic interactions in the bound-bound transitions in H-like ions !

A. Surzhykov et al. PRL 88 (2002) 153001

Effective anisotropy parameter: Multipole contributions

$$W(\theta) \propto 1 + \beta_{eff} P_2(\cos\theta)$$

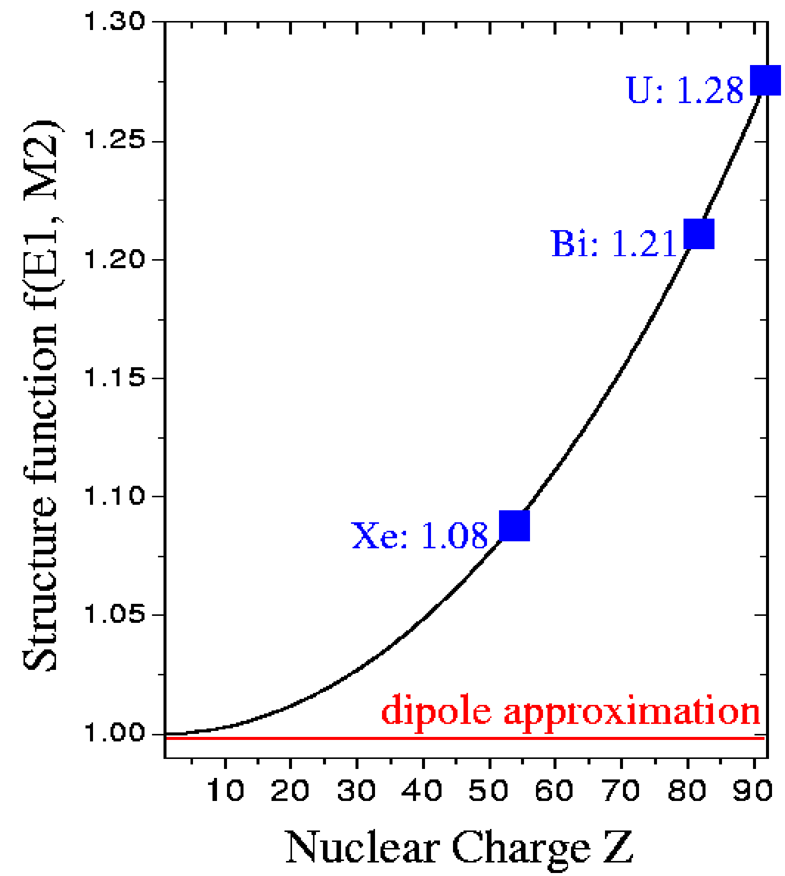
effective anisotropy parameter

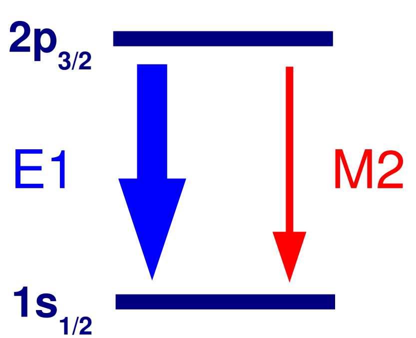
$$\beta_{eff} = \frac{1}{2} \underbrace{\frac{\sigma(\pm 3/2) - \sigma(\pm 1/2)}{\sigma(\pm 3/2) + \sigma(\pm 1/2)}}_{\text{alignment parameter (capture process)}} * \underbrace{f(E1, M2)}_{\text{structure function (ion)}}$$

alignment parameter (capture process)

structure function (ion)

$$f(E1, M2) \propto 1 + 2\sqrt{3}\frac{\langle |M2| \rangle}{\langle |E1| \rangle}$$





In contrast, contributions to decay rates appear additive:

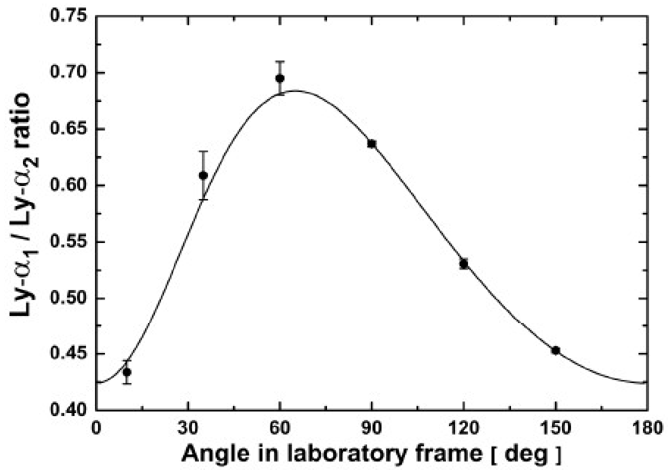
$$\frac{\Gamma_{M2}}{\Gamma_{tot}} \propto \frac{|\langle |M2| \rangle|^2}{|\langle |E1| \rangle|^2} \propto 0.008 \; !$$

even for U^{91+}

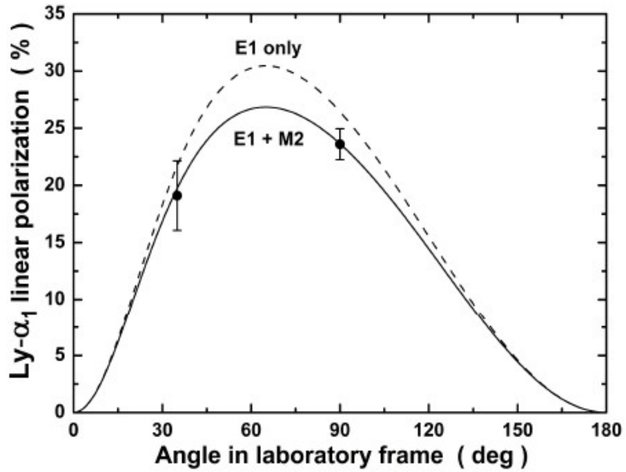
Elementary processes in strong Coulomb fields

– How can one directly ``measure'' multipole fields ?

Lyman-α_1 ($2p_{3/2}$ --> $1s_{1/2}$) for H-like U^{91+} ions:



Angular distribution



Linear polarization

$$W(\theta) \propto 1 + \beta_{20}^{\mathrm{eff}} \left(1 - \frac{3}{2} \sin^2 \theta \right)$$

$$P(\theta) = \frac{-\frac{3}{2} \gamma_{20}^{\mathrm{eff}} \sin^2 \theta}{1 + \beta_{20}^{\mathrm{eff}} \left(1 - \frac{3}{2} \sin^2 \theta \right)}$$

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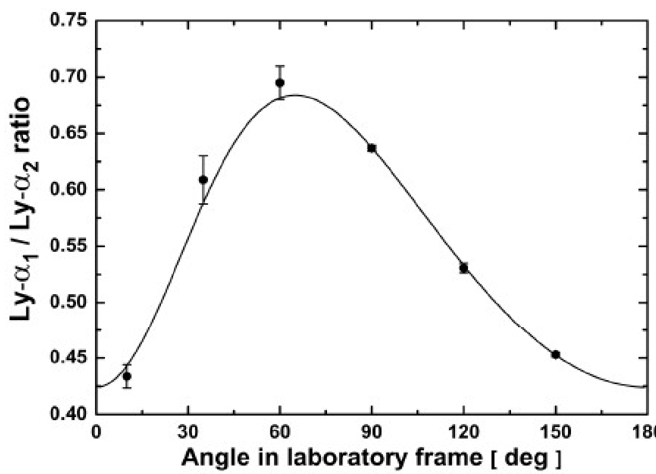
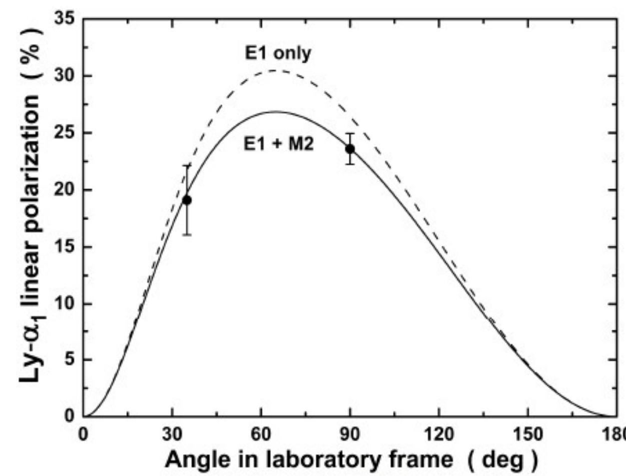


Angular distribution



Linear polarization

$$W(\theta) \propto 1 + \beta_{20}^{\mathrm{eff}} \left(1 - \frac{3}{2}\sin^2\theta\right)$$

$$f\,(A_2,\ a_{M2}/a_{E1})$$

$$P(\theta) = \frac{-\frac{3}{2}\gamma_{20}^{\mathrm{eff}} \sin^2\theta}{1 + \beta_{20}^{\mathrm{eff}}\left(1 - \frac{3}{2}\sin^2\theta\right)}$$

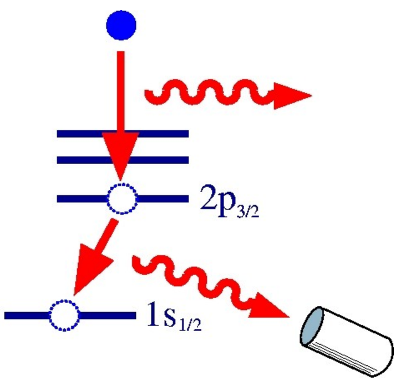
Alignment parameter $\mathcal{A}_2$		Amplitude ratio $a_{\mathrm{M2}}/a_{\mathrm{E1}}$	
Experiment	Theory	Experiment	Theory
-0.451 $\pm$ 0.017	-0.457	0.083 $\pm$ 0.014	0.0844

G. Weber et al., PRL 105 (2010) 243002.

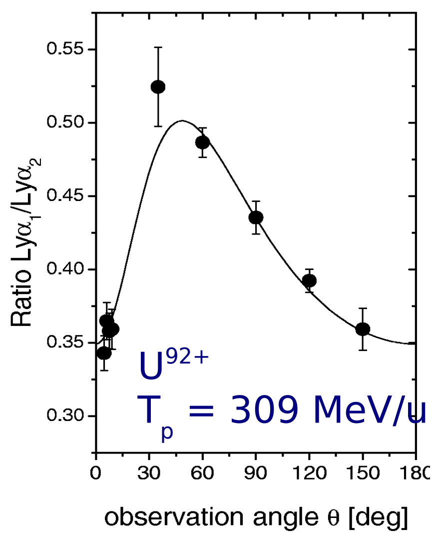
→ Model-independent and precise determination of the alignment and amplitude ratio.

Details matter: Adding one electron

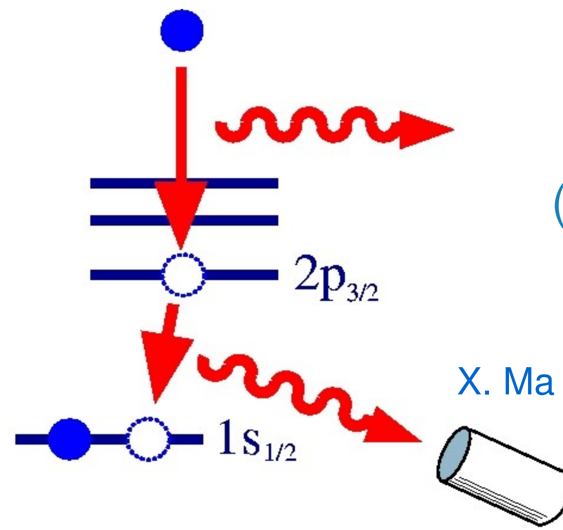
-- Lyman-α vs. K-α emission from high-Z ions



(initially) bare ion

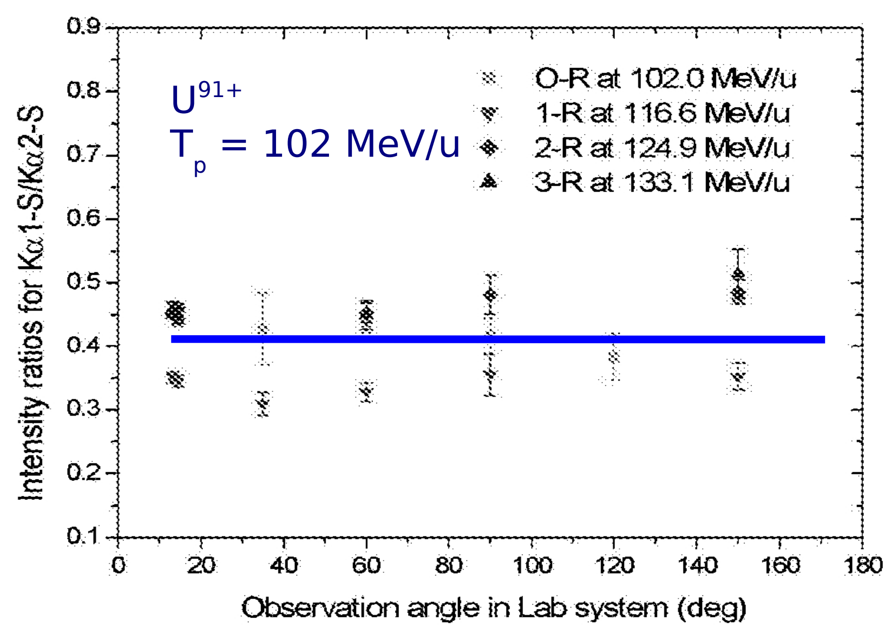


Ly-α_1 is strongly anisotropic



(initially) H-like ion

X. Ma et al, PRA 68 (2003) 042712.



K-α_1 is isotropic

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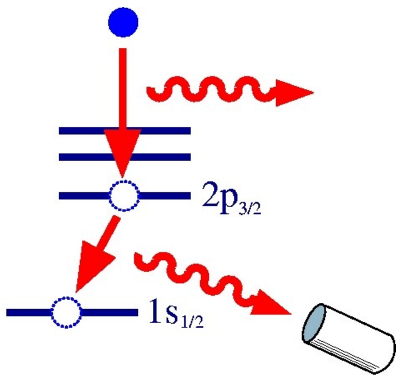

(initially) bare ion

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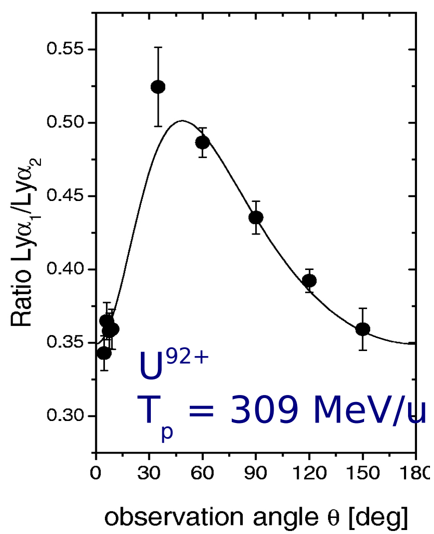

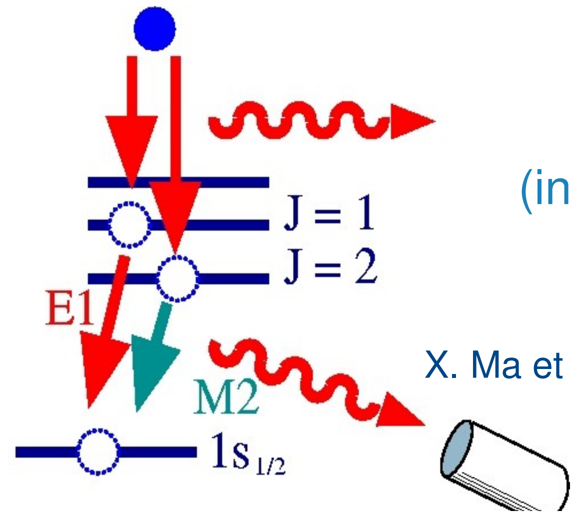

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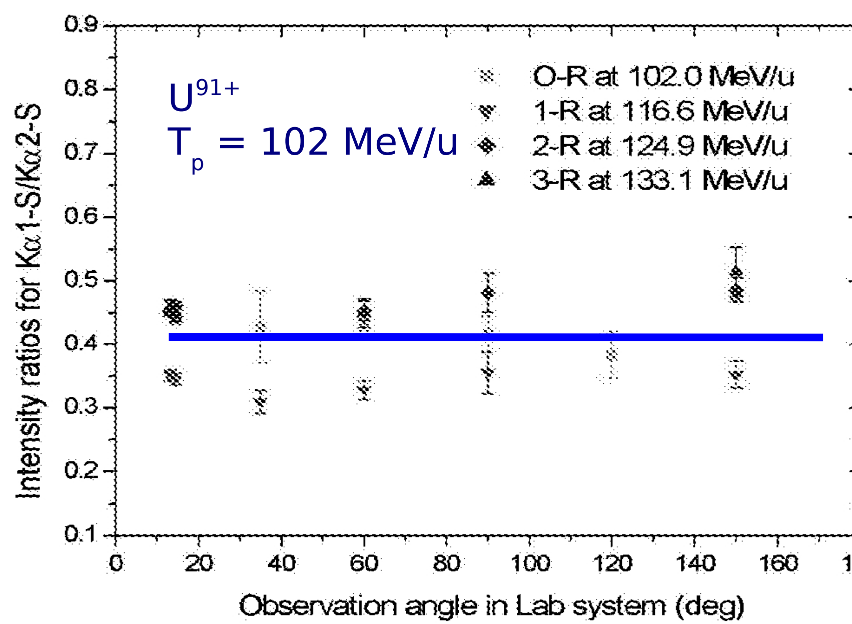


K-α_1 is isotropic

E1: $$W(\theta)_{E1} \sim 1 + \frac{1}{\sqrt{2}} A_2(J=1) P_2(\cos\theta)$$

M2: $$W(\theta)_{M2} \sim 1 - \sqrt{\frac{5}{14}} A_2(J=2) P_2(\cos\theta)$$

K-α decay of highly-charged ions

-- angular distribution as „observed" in experiments

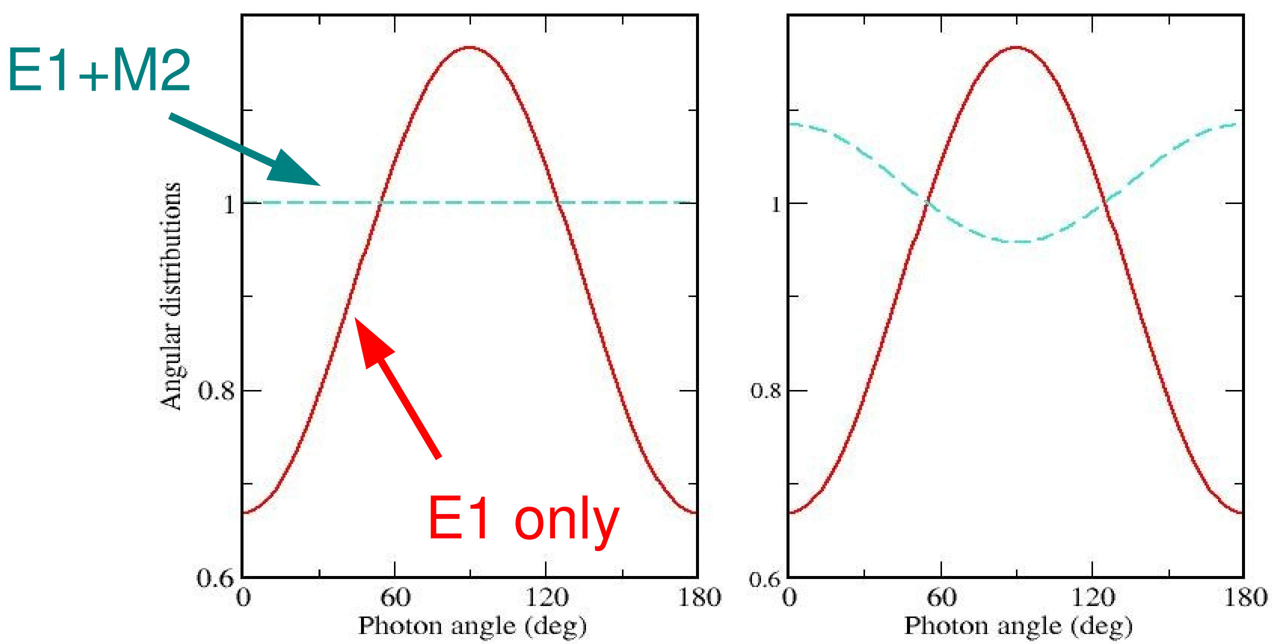
$$W(\theta)_{K\alpha_1} \sim N_{J=1} W_{E1}(\theta) + N_{J=2} W_{M2}(\theta)$$

A. Surzhykov et al., PRA 73 (2006) 032716.

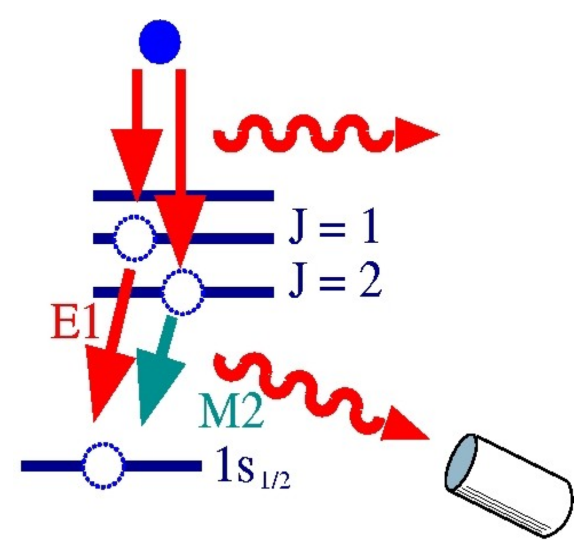
$$= 1 + \left(N_{J=1} \frac{1}{\sqrt{2}} A_2(J=1) - N_{J=2} \sqrt{\frac{5}{14}} A_2(J=2) \right) P_2(\cos\theta)$$

$N_{J=1}, N_{J=2}$ relative populations of J=1, 2 states

$N_{J=1} = N_{J=2} = \frac{1}{2}$ $\qquad$ $N_{J=1} = \frac{3}{8} \quad N_{J=2} = \frac{5}{8}$



Calculations have been done for L-REC of U^{91+} with T_p = 100 MeV/u



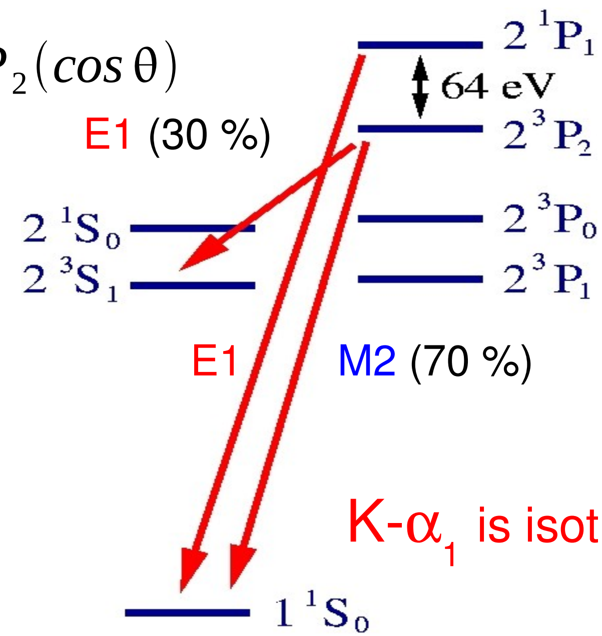
K-α decay of highly-charged ions

-- for 220 MeV/u U^{90+} ions following REC

$$W(\theta)_{K\alpha_1} \sim N_{J=1} W_{E1}(\theta) + N_{J=2} W_{M2}(\theta)$$

$$= 1 + \left(N_{J=1} \frac{1}{\sqrt{2}} A_2(J=1) - N_{J=2} \sqrt{\frac{5}{14}} A_2(J=2)\right) P_2(\cos\theta)$$

A. Surzhykov et al., PRA 73 (2006) 032716.



K-α_1 is isotropic

- Relative populations of the J = 1, 2 levels following REC (IPM model):

$$\frac{N_{J=1}}{N_{J=2}} = \frac{3}{5}$$

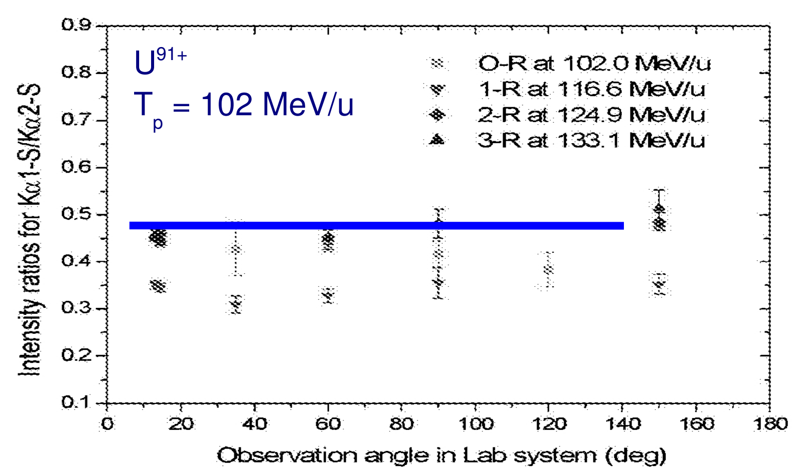
- By taking into account $^3P_2$ -> $^3S_1$ channel:

$$\frac{N_{J=1}}{N_{J=2}} = \frac{6}{7}$$

initial `capture' populations

\+ branching fractions

$$\boxed{\left(\frac{N_{J=1} - N_{J=2}}{N_{J=1} + N_{J=2}}\right)_{theory} \approx -0.08}$$



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- Properties of many-electron & (super-) heavy elements

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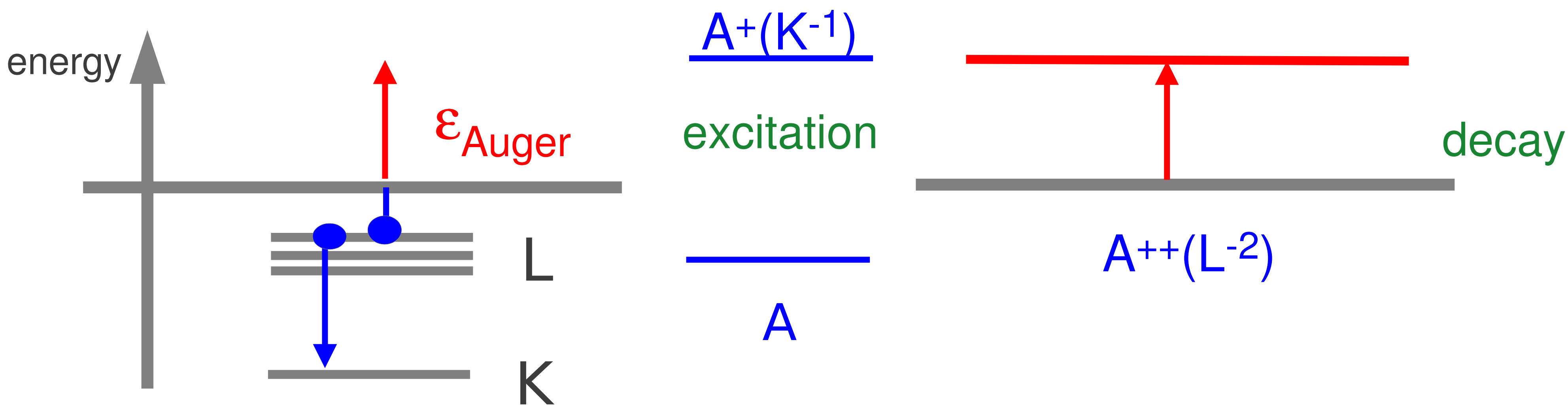
Auger emission from inner-shell excited states



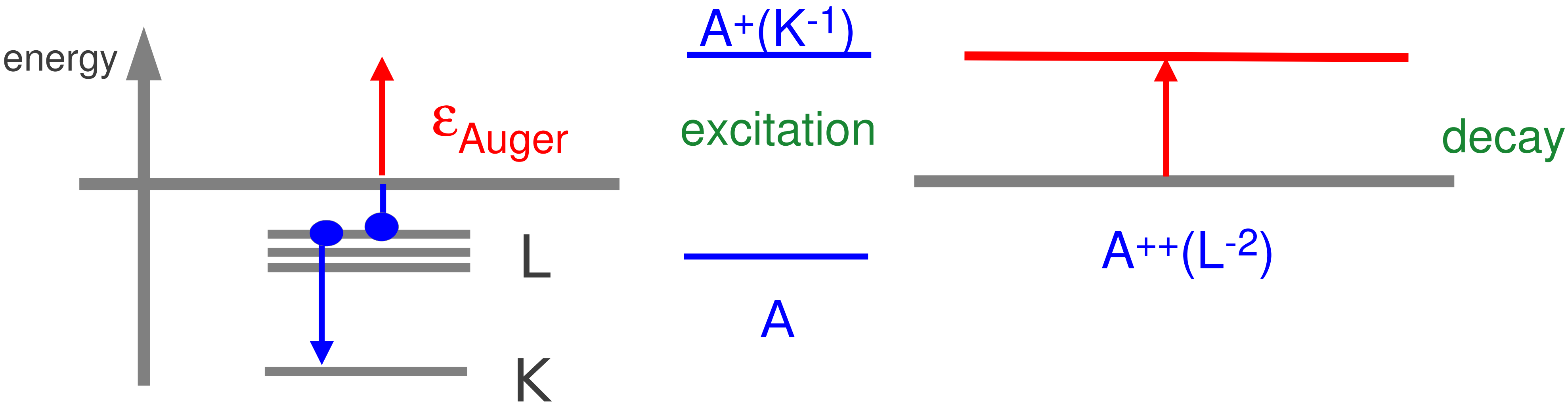
$$H = \sum_i \left(h_i + u(r_i)\right) \qquad H = \sum_i h_i + \sum_{i<j} \frac{1}{r_{ij}}$$

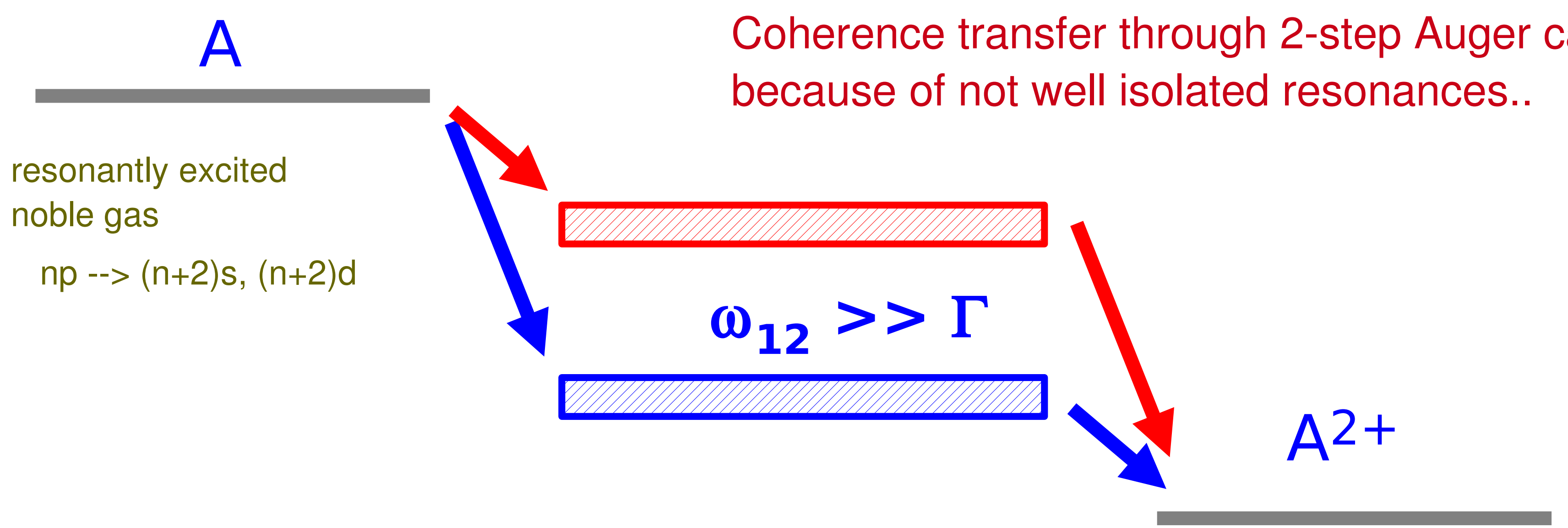
Wentzel's ansatz: Autoionization is caused by electron-electron interactions which cannot be considered in an one-particle picture.

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

Ideal tool for a better understanding of electronic correlations !

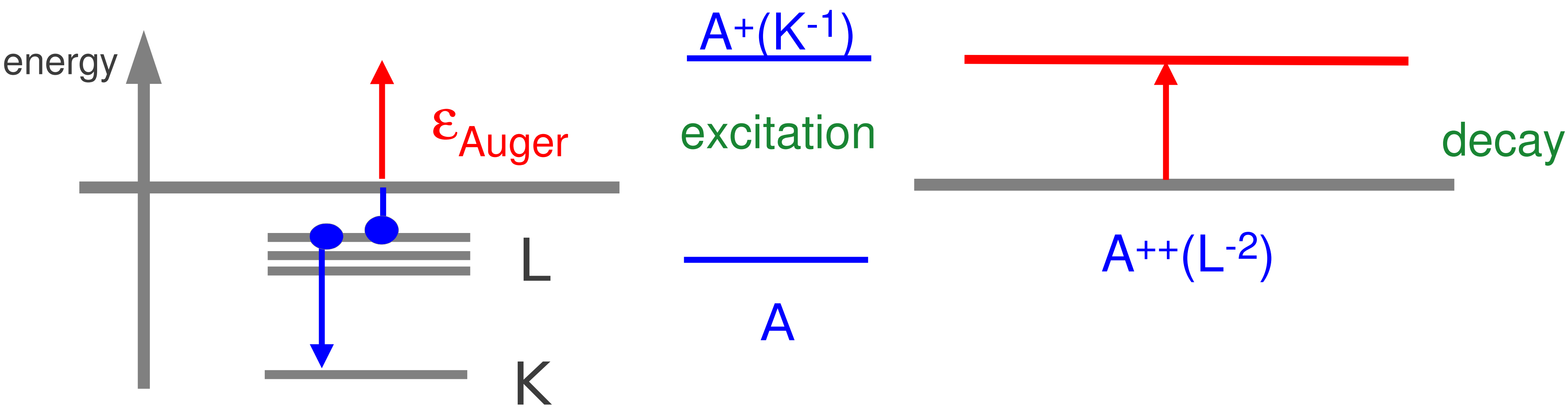
Auger emission from inner-shell excited states





Coherence transfer through 2-step Auger cascades
because of not well isolated resonances..

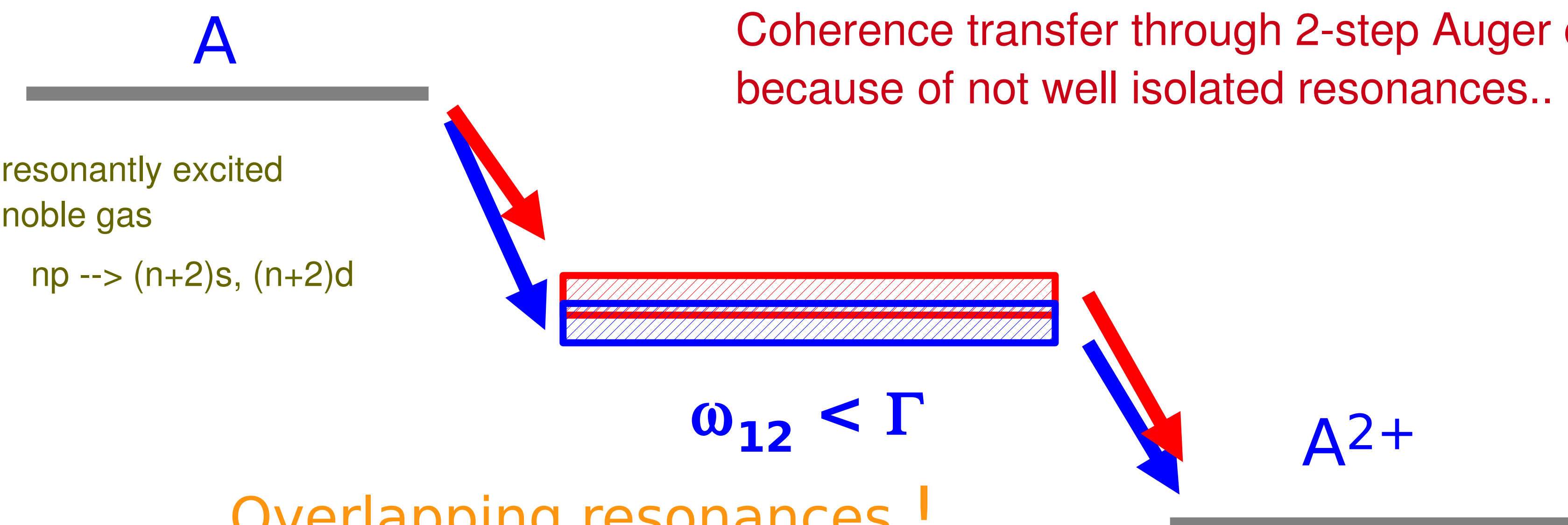
Auger emission from inner-shell excited states



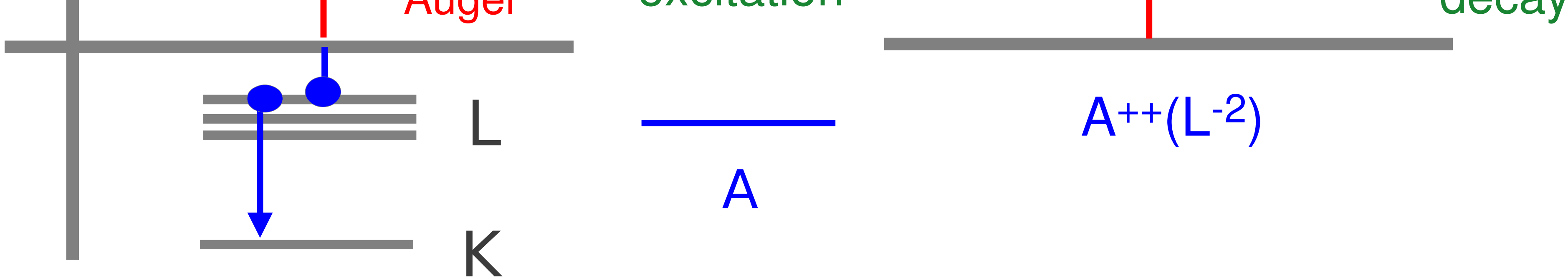
Coherence transfer through 2-step Auger cascades because of not well isolated resonances..

resonantly excited noble gas

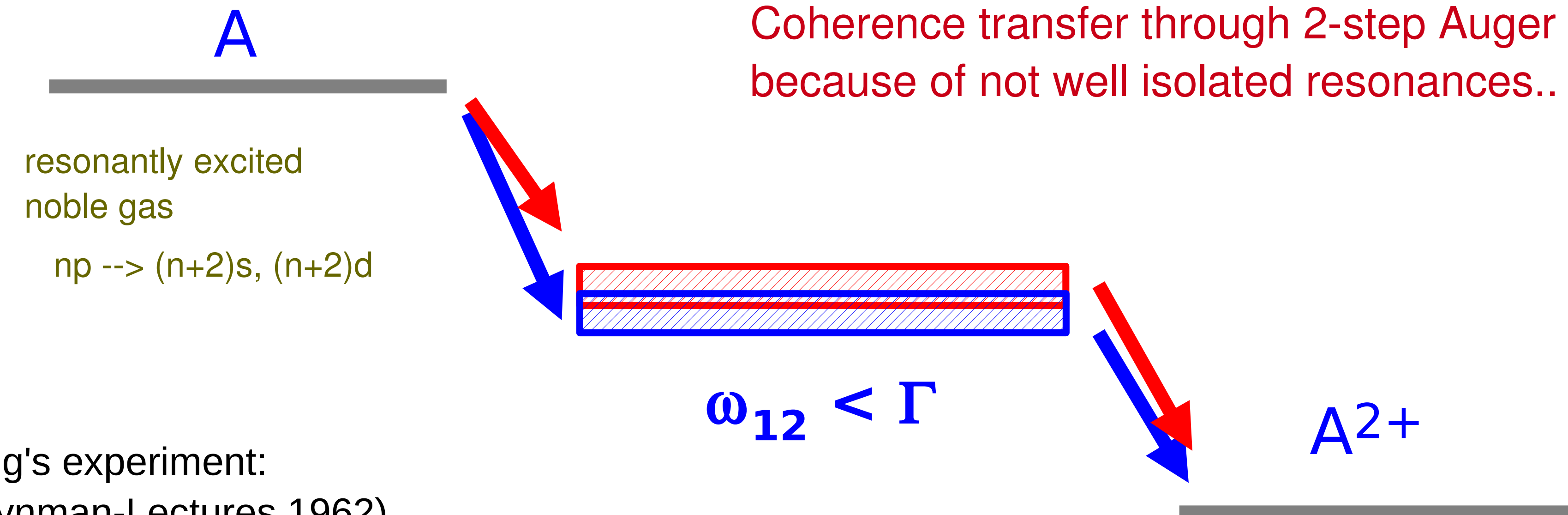
np --> (n+2)s, (n+2)d



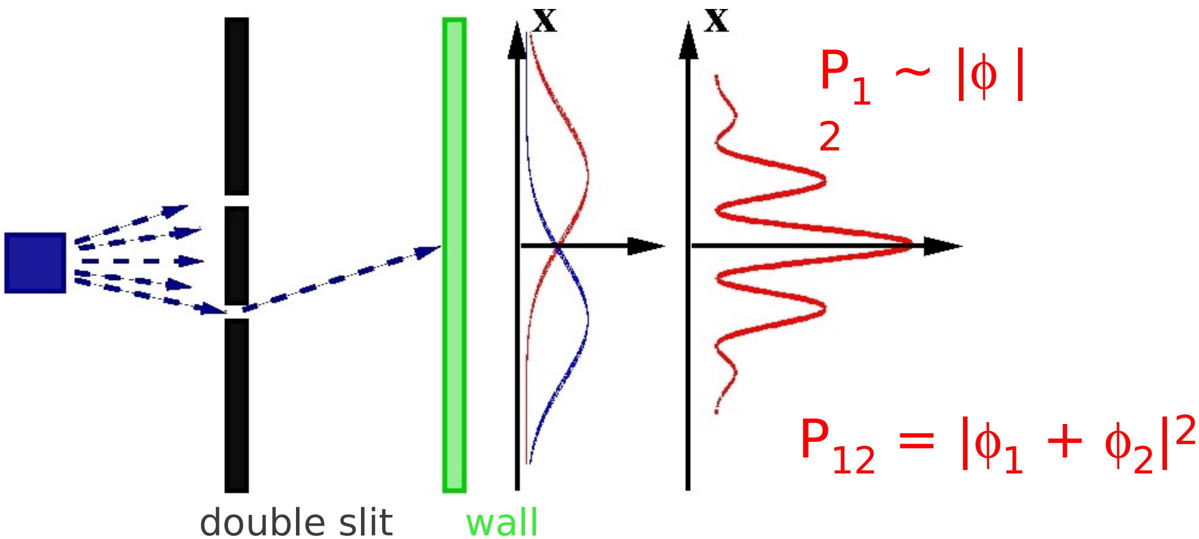
Overlapping resonances !



Coherence transfer through 2-step Auger cascades because of not well isolated resonances..

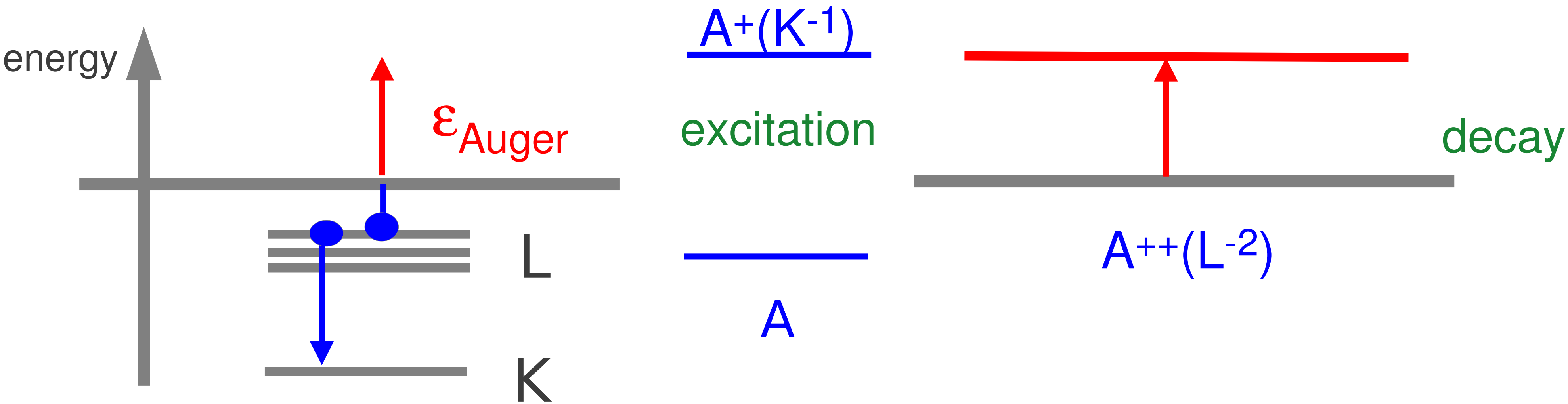


Young's experiment:
(Feynman-Lectures 1962)



How depend the Auger electron emission and, in particular, their angular distribution on the splitting of the resonances ?

Auger emission from inner-shell excited 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.

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

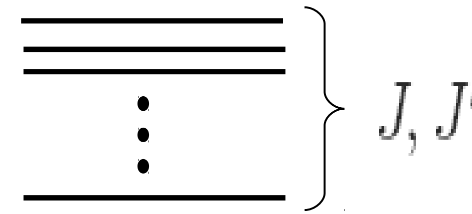
Breit interaction

Coupling of deeply-bound electrons to the continuum

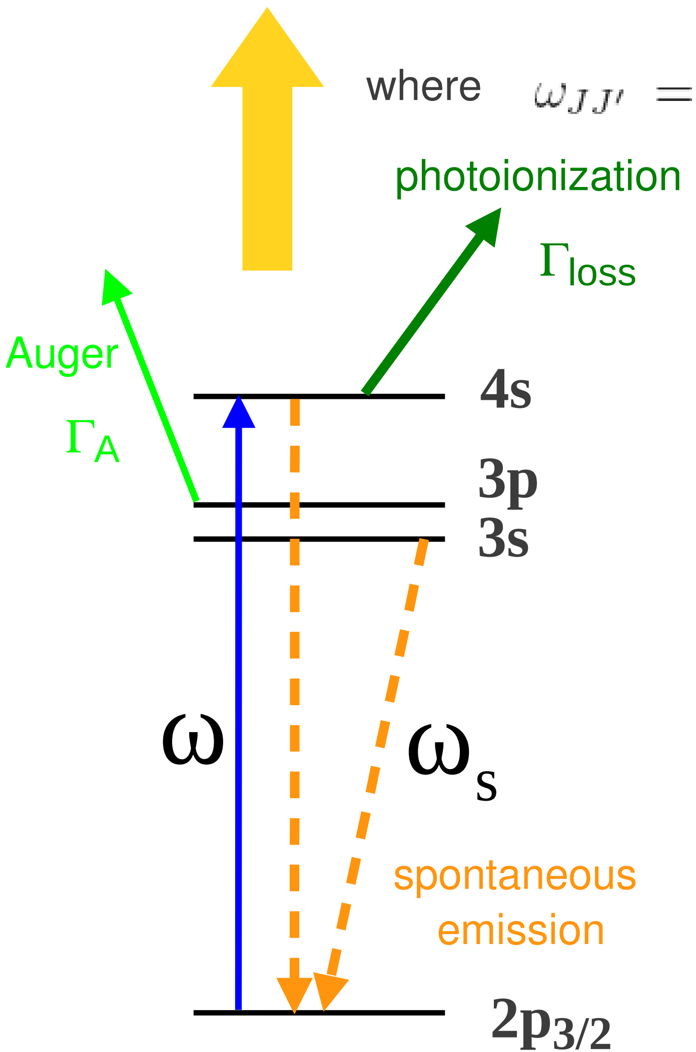
– correlation-induced autoionization and capture

Quantum evolution of the density operator:

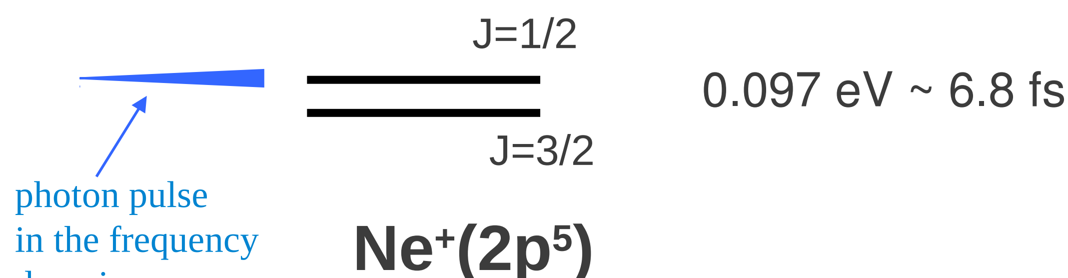
$$\rho_{kq}(J,J';t) = \rho_{kq}(J,J';t_1)\exp[(i\omega_{JJ'} - \Gamma_{JJ'})(t-t_1)]$$



where $\omega_{JJ'} = E_J - E_{J'} \quad \Gamma_{JJ'} = (\Gamma_J + \Gamma_{J'})/2$



For short pulses, the excitation occurs coherently:



Coupling of deeply-bound electrons to the continuum

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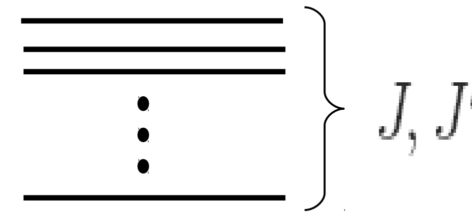
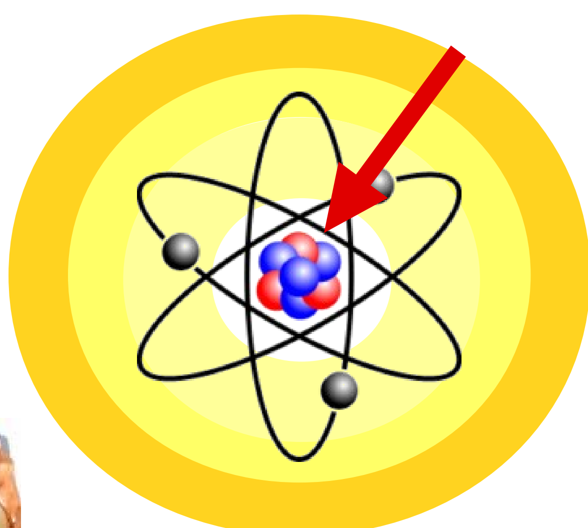
Quantum evolution of the density operator:



$$\rho_{kq}(J, J'; t) = \rho_{kq}(J, J'; t_1) \exp[(i\omega_{JJ'} - \Gamma_{JJ'})(t - t_1)]$$

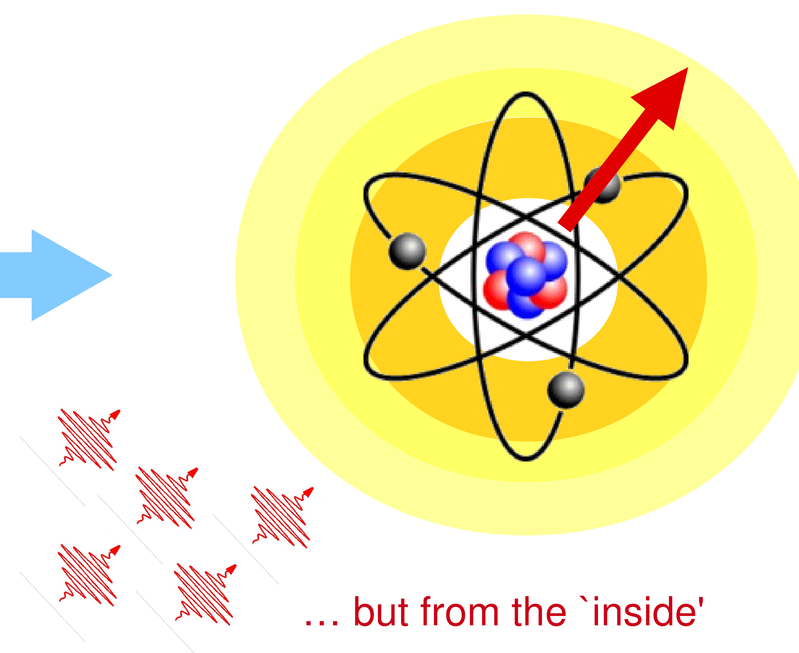
where $\omega_{JJ'} = E_J - E_{J'}$ $\Gamma_{JJ'} = (\Gamma_J + \Gamma_{J'})/2$

Intense optical and VUV laser



peels off the electrons layer by layer

intense FEL radiation



... but from the `inside'

Correlated quantum systems in intense fields

– dynamics of multiple and highly-charged ions

- Ionization & recombination in strong Coulomb fields

  correlated photon and/or electron emission

- Coupling of deeply-bound electrons to the continuum

  correlation-induced autoionization & capture

- Properties of many-electron & (super-) heavy elements

  correlated (ab-initio) many-body techniques

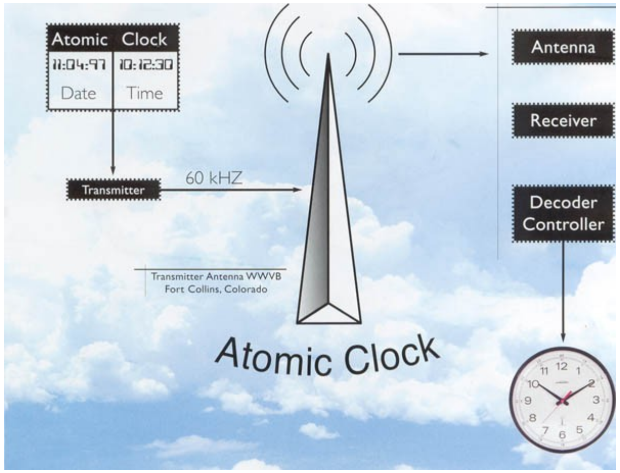
- Multi-photon & electron dynamics in intense FEL radiation

  correlated many-electron dynamics

Properties of many-electron & (super-) heavy elements

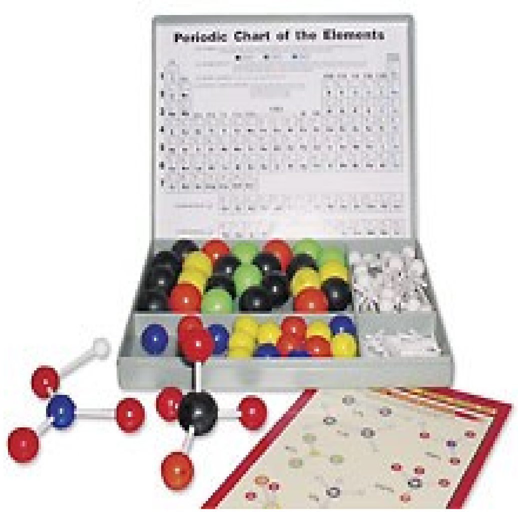
– there is a great need for accurate atomic amplitudes

- Analysis and interpretation of optical and x-ray spectra (astro physics)
- Diagnostics of astro physical and laboratory plasmas
- Development of UV/EUV light sources and lithograhpy
- Frequency standards and atomic clocks
- Spectroscopy on heavy and superheavy elements (actinides, transactinides)
- Isotope shifts and hyperfine structures
- Nonradiative (inner-shell) transitions and autoionization
- Ion recombination and photon emission
- Multi-photon processes
- …
- ...
- „Complete experiments“
- Parity nonconservation (PNC)
- Search for electric dipole moments



Different `systematic' ab-initio approaches exist

– to describe the electronic structure of atoms and ions



Multiconfiguration expansions

$$\psi_\alpha(P J M) = \sum_{r}^{n_c} c_r(\alpha)\, \gamma_r P J M >$$

Construct a `physically motivated' basis
in the N-electron Hilbert space.

Many-particle character
„electronic correlations“

$\longleftrightarrow$

Relativistic effects

Shell structure
static vs. dynamic correlations

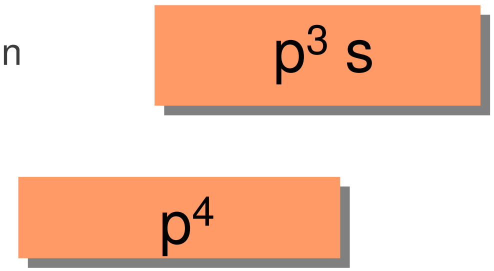
Direct vs. indirect effects
QED corrections

Generalization of the knowledge about (Dirac's) one- or few-electron atoms
in such a way to enable the „computation“ of heavy atoms and ions.

Electronic correlations: A simple man's view

Fine-structure of open-shell atoms and ions

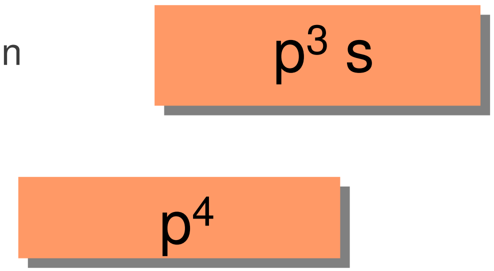
	No. of levels for single configuration
p^4	5
$p^3\,s$	10



Electronic correlations: A simple man's view

Fine-structure of open-shell atoms and ions

	No. of levels for single configuration
p^4	5
$p^3\ s$	10
d^8	9
$d^7\ p$	110



Electronic correlations: A simple man's view

Fine-structure of open-shell atoms and ions

	No. of levels for single configuration
p^4	5
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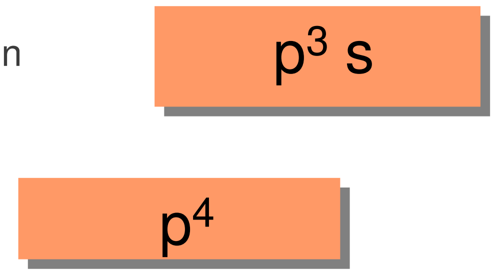
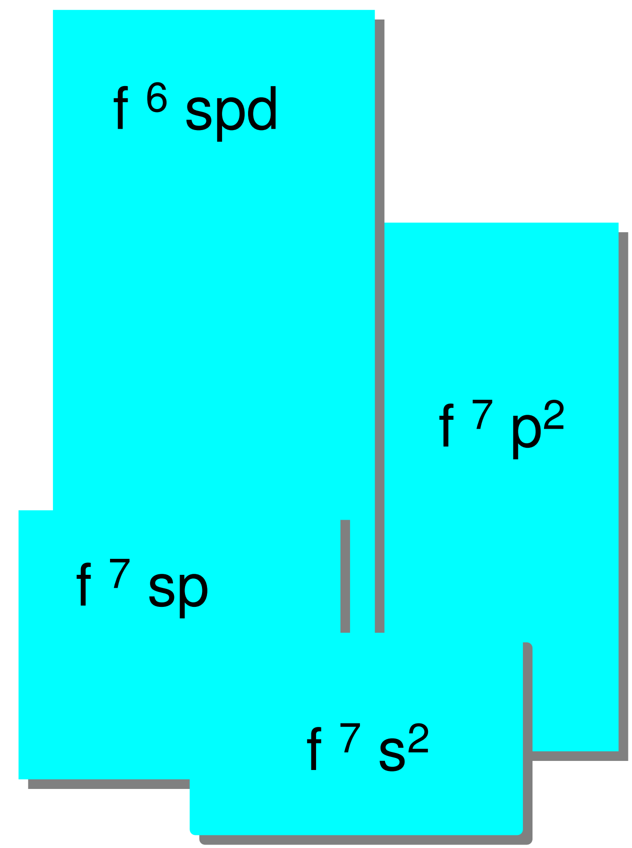
$f^{\,7}\,s^2$	327
$f^{\,7}\,sp$	3808
$f^{\,7}\,p^2$	4724
$f^{\,6}\,spd$	31804



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



Electronic correlations: A simple man's view

Fine-structure of open-shell atoms and ions

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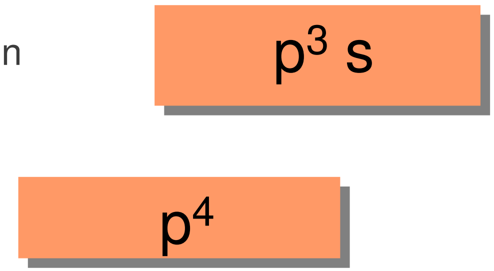
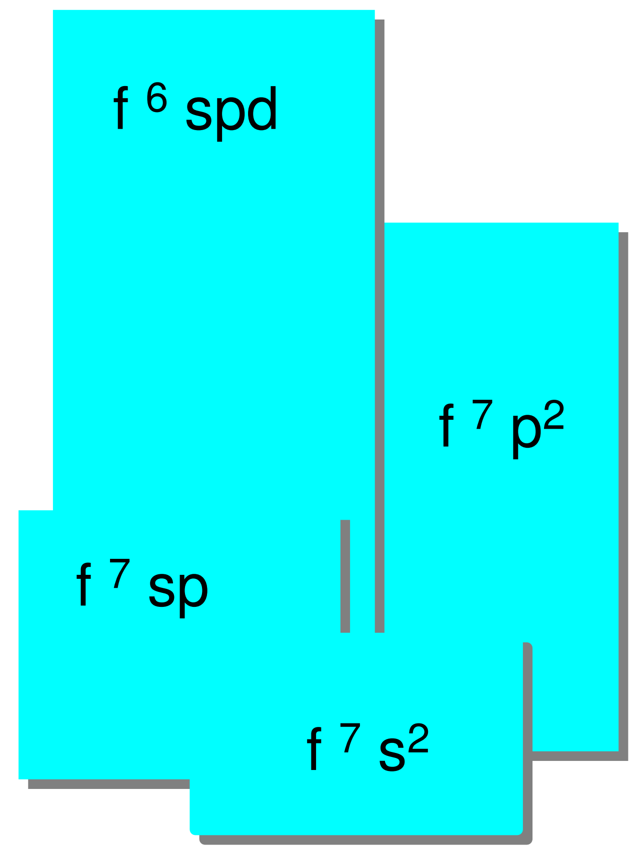


~ 0.1 .. 1 %

level and transition energies

~ 10 ... 30 %

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



Concept of atomic shells and electronic configurations gets lost for heavy open-shell systems !

Electronic correlations: A simple man's view

Fine-structure of open- … ns

No. of levels
for single configuration

p^4	5
$p^3\ s$	10
d^8	9
$d^7\ p$	110
$f^7\ s^2$	
$f^7\ s$…	

$p^3\ s$

p^4

… ents depend
… e shell structure
… oms and ions !

… sition
… rgies

~ 10 ... 30 %

f^6 spd

$f^7\ p^2$

f^7 sp

$f^7\ s^2$

Conce… e shells and electronic configurations
gets lost … eavy open-shell systems !

There is no simple path from one element to the next,
from one ionization state to the other,
from one property to ...

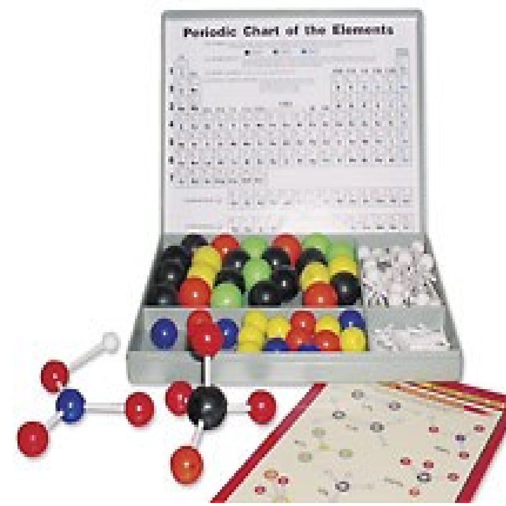
Different `systematic' ab-initio approaches exist

– to describe the electronic structure of atoms and ions

Multiconfiguration expansions

$$\psi_\alpha(PJM) = \sum_{r}^{n_c} c_r(\alpha)\, \gamma_r P J M >$$

Construct a `physically motivated' basis
in the N-electron Hilbert space.



Many-particle character
„electronic correlations“

Shell structure
static vs. dynamic correlations

Generalization of the knowledg
in such a way to enable the „co

Energies & Wave functions

- Cowan / CIV3
- MCHF
- GRASP(-92) / RATIP
- „Desclaux“
- Coupled-Cluster

S. Fritzsche, JESRP 114-116 (2001) 1155; Comput. Phys. Commun. 183 (2012) 1525

RATIP

Relativistic Atomic Transition, Ionization and Recombination Properties

AUGER: Auger rates, relative intensities, angular distribution & spin polarization parameters.

REC: Radiative recombination & electron capture rates, angular parameters.

CESD: Determinant representation of atomic and configuration state functions.

RELCI: Relativistic configuration interaction wave functions & QED estimates.

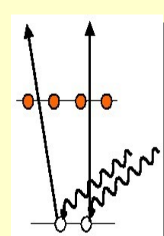
REOS: Relaxed-orbital Einstein A and B coefficients, transition probabilities and lifetimes.

TOOLBOX: Level energies and notations; manipuations of file interfaces, miscelaneous.

COULOMB: Exitation amplitudes, (M_J-dependent) cross sections, alignment parameters.

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

S. Fritzsche, JESRP 114-116 (2001) 1155; Comput. Phys. Commun. 183 (2012) 1525

RATIP

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REOS: Relaxed-orbital Einstein A and B coefficients, transition probabilities and lifetimes.

PHOTO: Ionization cross sections, angular & spin-polarization, alignment of photoions.

TOOLBOX: Level energies and notations; manipuations of file interfaces, miscelaneous.

ANCO: Angular coefficients for scalar and non-scalar operators.

COULOMB: Exitation amplitudes, (M_J-dependent) cross sections, alignment parameters.

S. Fritzsche, JESRP 114-116 (2001) 1155; Comput. Phys. Commun. 183 (2012) 1525

S. Fritzsche, JESRP 114-116 (2001) 1155; Comput. Phys. Com... 183 (2012) 1525

M. Sewtz, M. Block et al., private commun. (2007 and later)

Experimental proposal: Optical spectroscopy of nobelium (Z=102)

-- on-line production of target atoms

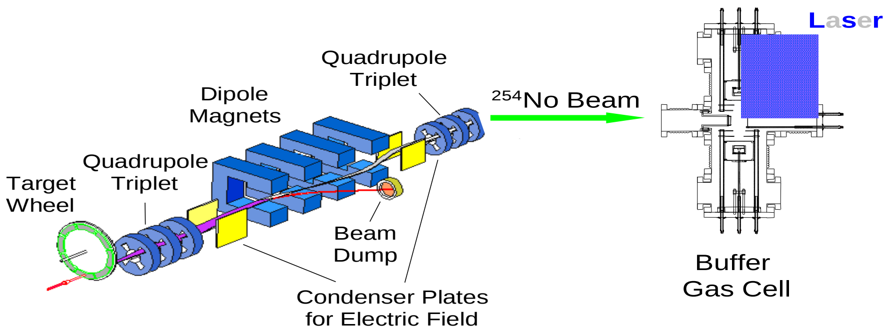
Ground-state configuration:

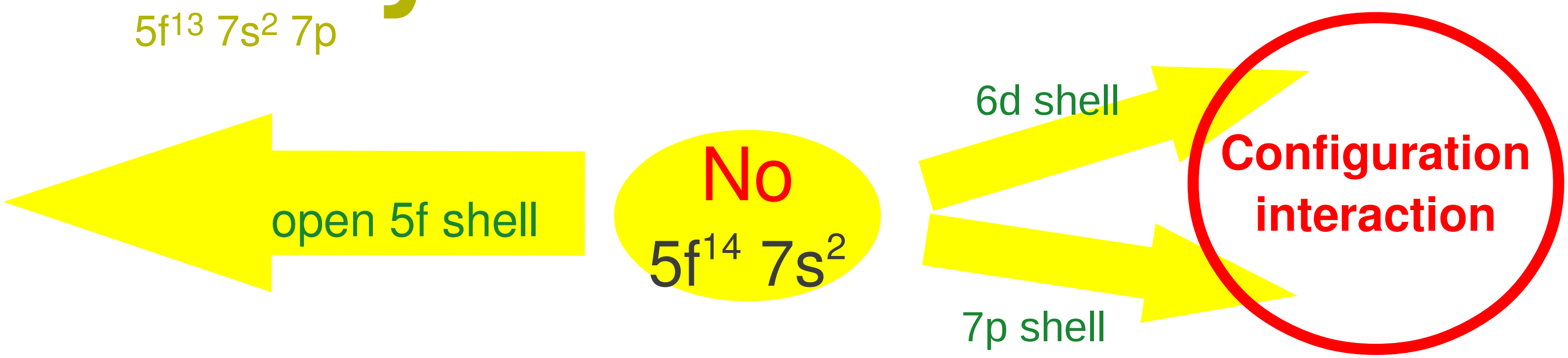
[Rn] $5f^{14}\,7s^2$

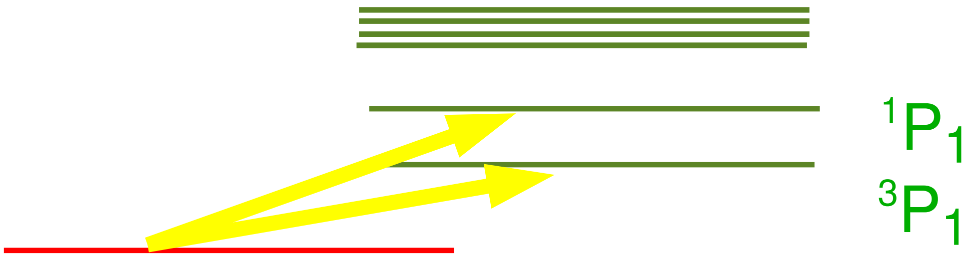
Low-lying excitations:

[Rn] $5f^{14}\,7s\,7p$

$\left.\begin{array}{l} 5f^{13}\,6d\,7s^2 \\ 5f^{14}\,6d\,7s \\ 5f^{14}\,7s\,8s \\ 5f^{13}\,7s^2\,7p \end{array}\right\}$ 2 ... 5 eV







	Level	$^{2S+1}L_J$	Excitation energy (eV) Model I	Model II
No	$5f^{14}7s7p$	$^3P_1$	2.34	2.60
	$5f^{14}7s7p$	$^1P_1$	3.49	3.36

S. Fritzsche, EJP D33, 15 (2005)

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

Calculation of low-lying levels for (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

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

Calculation of low-lying levels for (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 ($n = 6$).

	$^2D_{3/2} - {}^2P^o_{1/2}$			$^2D_{3/2} - {}^2P^o_{3/2}$		
Expansion	gf_L	gf_V	Scaled gf_L	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	

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

Calculation of low-lying levels for (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 ($n = 6$).

	$^2D_{3/2} - {}^2P^o_{1/2}$			$^2D_{3/2} - {}^2P^o_{3/2}$		
Expansion	gf_L	gf_V	Scaled gf_L	gf_L	gf_V	Scaled gf_L
Lu						
VV + CV($4f^{14}$)	0.0304	0.0582	0.0315	0.0111	0.0219	0.0114
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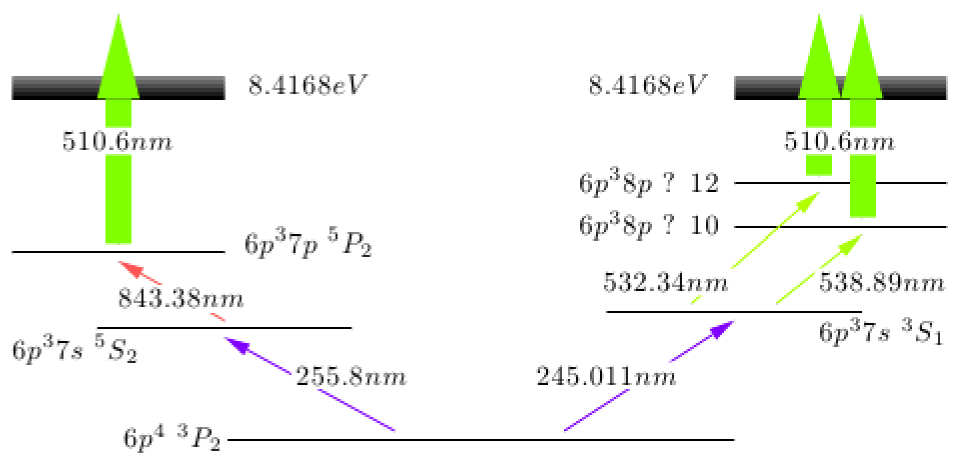
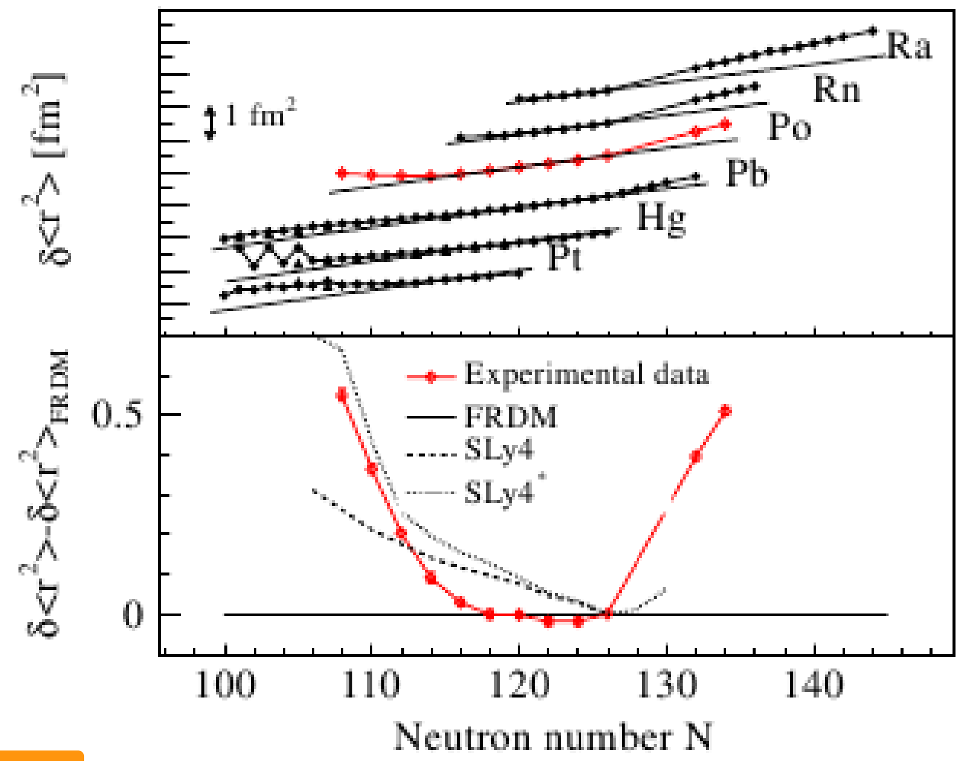
Good accuracy of the (atomic) energies is
a necessary, but not a sufficient criterion !

Po^+ (Z=84): Heavy ions with complex spectra

-- the challenge of excited states and half-filled shells

$6p^37s\ ^5S_2 \rightarrow 6p^37p\ ^5P_2$





Transition [nm]	F [GHz/fm^2]	M_{SMS} [GHz $\cdot$ amu]
255.8	28.363	51
843.38	−12.786	−311

T. Cocolios et al., Phys. Rev. Lett. 106 (2011) 052503.

Further isotopic chains

-- current requests from laser spectroscopy

Ion (Z)		Transition	λ [nm]	M [GHz·amu]	F [MHz/ fm^2]	Method & Ref.
Sc^+	(21)	$3d4s\ ^3D_2 - 3d4p\ ^3F_3$	363.1	580 (10%)	-355 (15%)	MCDF (Avgoulea *et al.* 2011)
Mn^+	(25)	$3d^54s\ ^5S_2 - 3d^54p\ ^5P_3$	295	-572 (15%)	852 (10%)	MCDF (Charlwood *et al.* 2010)
Cu^+	(29)	$3d^{10}4s\ ^2S_{1/2} - 3d^{10}4p\ ^2P_{3/2}$	325	... (15%)	-680 (15%)	MCDF
Ga^+	(31)	$4p\ ^2P_{3/2} - 5s\ ^2S_{1/2}$	417	-534 (15%)	392 (10%)	MCDF
Y^+	(39)	$5s^2\ ^1S_0 - 4d5p\ ^1P_1$	363	1789[a]	-3181[a]	semi-emp.
				1318 (15%)	-3210 (10%)	MCDF, this w.
		$4d5s\ ^3D_2 - 4d5p\ ^3P_1$	321	124 (10%)	-1132 (10%)	MCDF, this w.
Os^-	(76)	$5d^76s^2\ ^4F_{9/2} - 5d^66s^26p\ ^6D_{9/2}$	1162.7	2,500 ± 12,600	16,200 ± 9,900	(Kellerbauer *et al.* 2011)[b]
				4,000 (40%)	12,300 (25%)	MCDF
Po^+	(84)	$6p^37s\ ^5S_{1/2} - 6p^37p\ ^5P_2$	843.4	-311 (15%)	-12,786 (15%)	MCDF

[a] Semi-empirical estimates by using radii from several neighbored elements; see text for discussion.
[b] Experiments have been performed for two stable isotopes $^{187}Os^-$ and $^{189}Os^-$ by Kellerbauer *et al.* (2011).

Conclusions:

- Use transitions between low-lying levels with stable electronic structure (filled shells)
- If possible, use some charge state “near to” closed-shell (ground-state) structures

Correlated quantum systems in intense fields

– dynamics of multiple and highly-charged ions

- Ionization & recombination in strong Coulomb fields

  correlated photon and/or electron emission

- Coupling of deeply-bound electrons to the continuum

  correlation-induced autoionization & capture

- Properties of many-electron & (super-) heavy elements

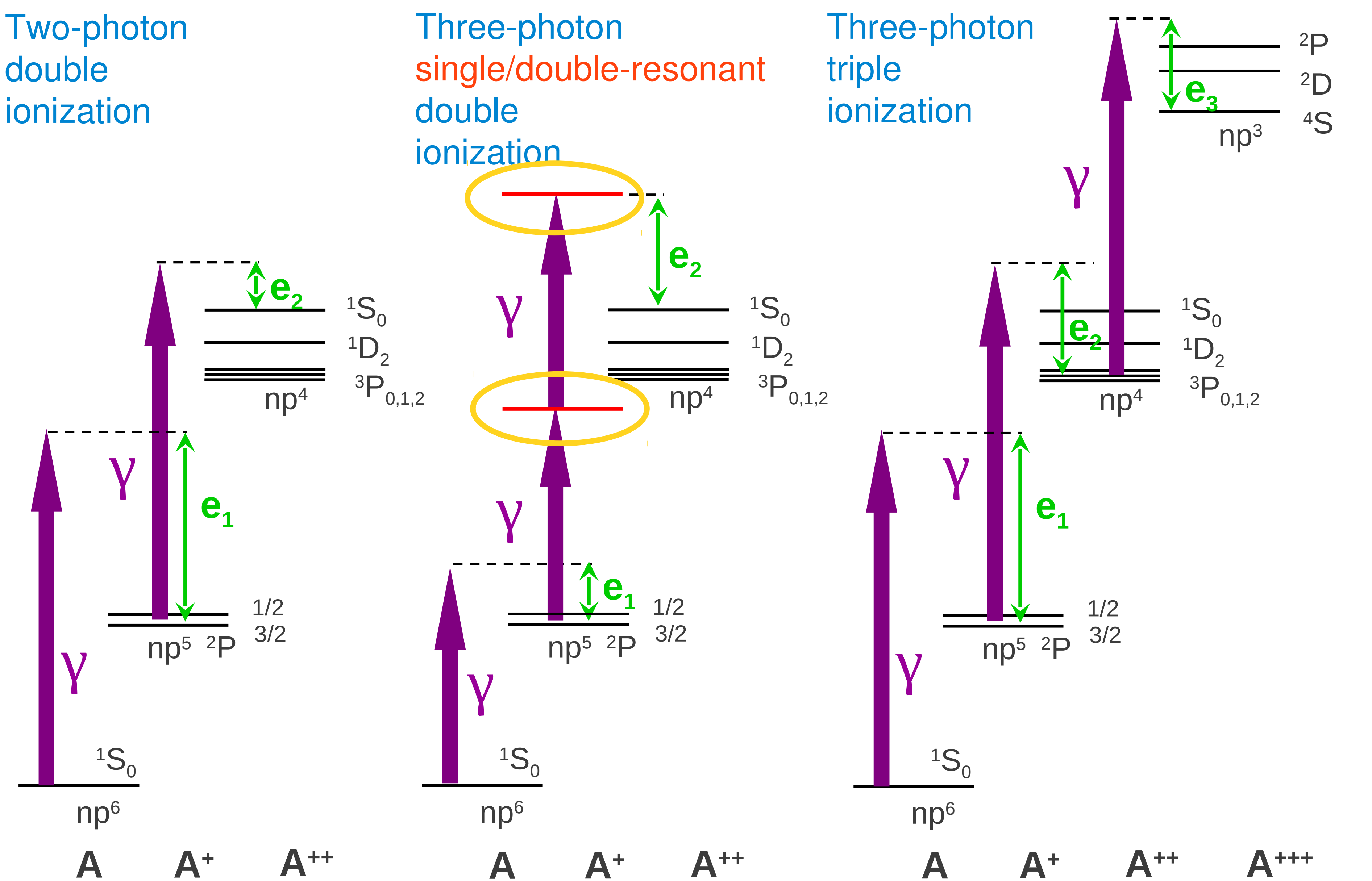
  correlated (ab-initio) many-body techniques

- Multi-photon & electron dynamics in intense FEL radiation

  correlated many-electron dynamics

Multi-photon & electron dynamics in intense FEL radiation

– correlated many-electron dynamics



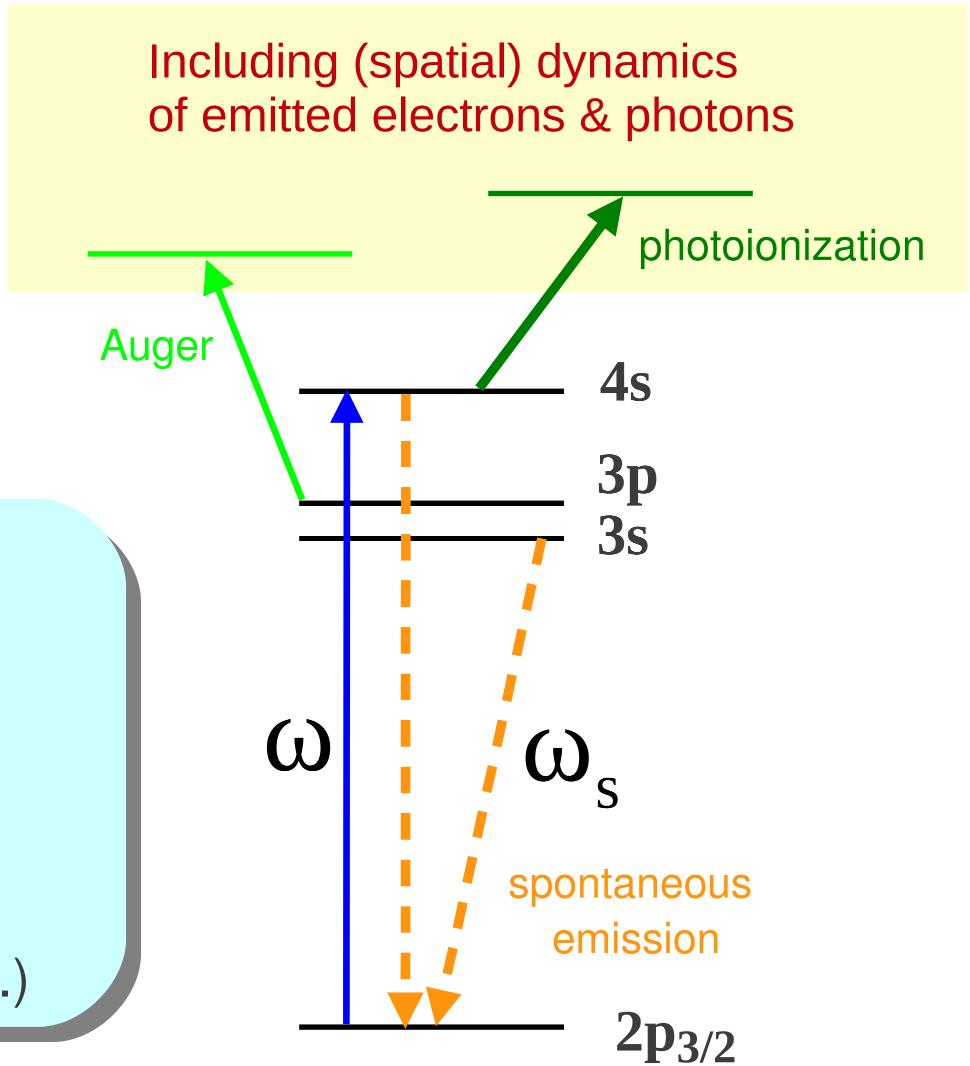
Multi-photon & electron dynamics in intense FEL radiation

– coupling to the Auger continuum

Explicitly time-dependent density operator:

$$\dot{\rho} \;=\; \frac{i}{\hbar}\,[H,\,\rho] \;+\; L\,\rho$$

Based on Liouville's equation



Frequently applied in quantum optics:

- N (non-degenerate) bound levels
- long pulses with linear polarization
- electric-dipole & RWA approximation
- influence of coupling fields

(EIT, slow light, ...)

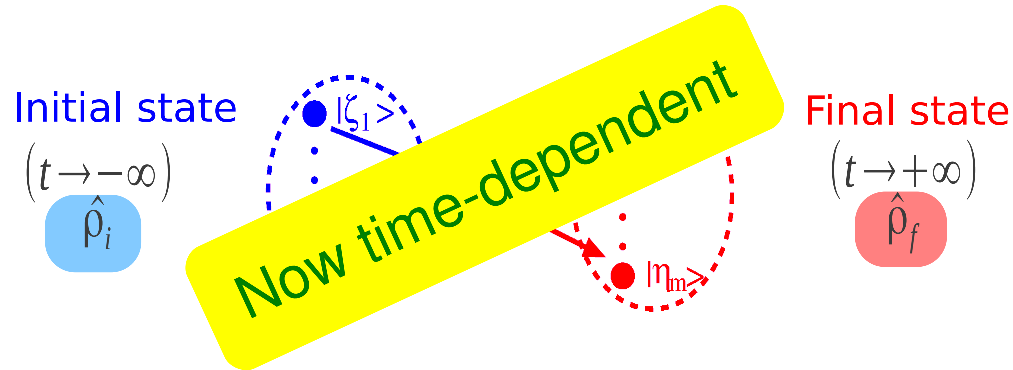
Coherent time evolution of inner-shell excited systems

– in short-pulse or pump-probe experiments

Explicitly time-dependent density operator including spatial degrees of freedom:

$$\dot{\rho} = \frac{i}{\hbar} [H, \rho] + L\,\rho$$

Based Liouville's equation



$$i \frac{d\,\rho_{kq}(\alpha,\beta)}{dt} = \sum_{\kappa' q'} \sum_{\gamma} \Big\{ F_{\kappa q}^{\kappa' q'} \left(\alpha,\beta,\gamma, \begin{array}{c}\text{pulses}\\ \text{geometry}\end{array}; t\right) \rho_{k'q'}(\gamma,\beta)$$

direct coupling

$$- G_{\kappa q}^{\kappa' q'} \left(\alpha,\beta,\gamma, \begin{array}{c}\text{pulses}\\ \text{geometry}\end{array}; t\right) \rho_{k'q'}(\alpha,\gamma)$$

exchange

$$- i\,\Gamma_{\kappa q}^{\kappa' q'} (\alpha,\beta,\gamma; t)\, \rho_{k'q'}(\gamma,\beta) \Big\}$$

ionization & loss processes

Atomic (transition & ionization) amplitudes from many-body theory (RATIP)

S. Fritzsche, CPC 183 (2012) 1525

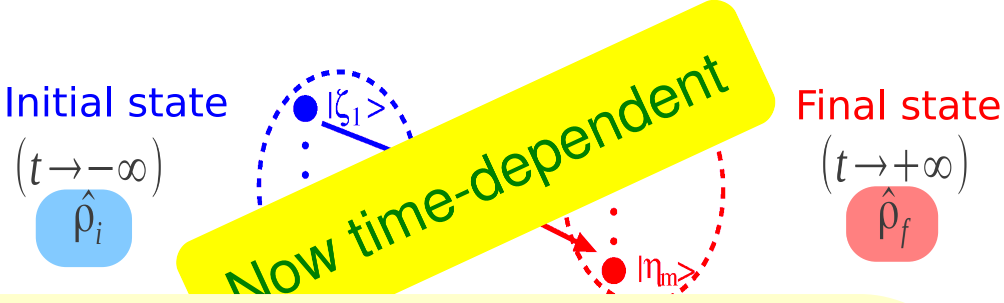
Collaboration with Alexei Grum-Grzhimailo

Coherent time evolution of inner-shell excited systems

– in short-pulse or pump-probe experiments

Explicitly time-dependent density operator including spatial degrees of freedom:

$$\dot{\rho} \;=\; \frac{i}{\hbar}\,[H,\,\rho] \;+\; L\,\rho$$



Based Liouville's equation

The "hope": Compared with time-dependent SE

- control of approximations: RWA, dipole, … based on "physical insight"
- explicit electron dynamics

  (including angular-and spin-dependent emission)
- pulses with given shape and polarization
- higher multipole contributions
- (rather) simple control upon effects of level and pulse structures
- feasibe for many-electron systems in 3D

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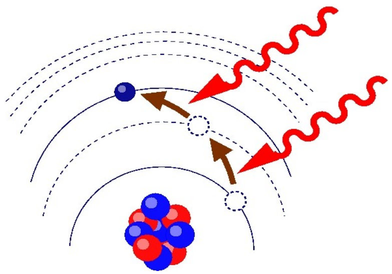
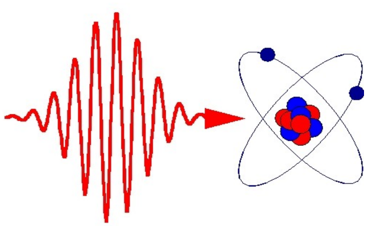
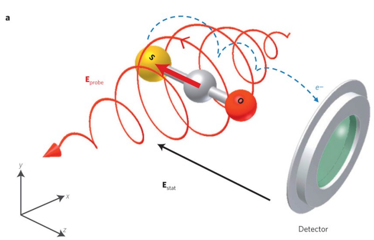
Collaborati

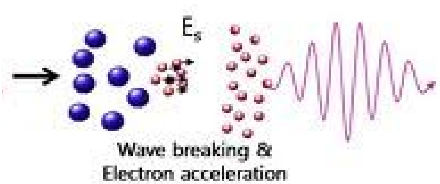
Quantum dynamics of correlated systems

– (time-) evolution “through” the density matrix









Density matrix

$$\rho \;=\; \rho(r,t;\, r',t')$$

$$\;=\; \rho(\mu_S,\, J,\, J';\, E;\, l,\, \mu_{l;}\, t)$$

Ensemble of (collision) systems: requires statistical description

- Can be used easily to accompany the system through several time-independent (or time-dependent) interactions, including the capture or emission of photons, electrons, etc. !

Summary: combination of quite different (many-particle) techniques

- Schrödinger equation
- Dirac equation
- Quantum electrodynamics
- Group theory
- Density matrix theory

Study of atomic processes and quantum dynamics

Combination of different concepts

Use of systematic approximations to describe the behaviour of quantum systems:

- Density matrix techniques and spherical tensors
- Racah's algebra
- Multiconfigurational expansions (CI, MCDF)
- Many-body perturbation theory (MBPT, CC)
- Green's functions